

Effective Hamiltonian for a half-filled asymmetric ionic Hubbard chain with alternating on-site interaction

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We derive an effective spin Hamiltonian for the one-dimensional half-filled asymmetric ionic Hubbard model (IHM) with alternating on-site interaction in the limit of strong repulsion. It is shown that the effective Hamiltonian is that of a spin $S = 1/2$ anisotropic XXZ Heisenberg chain with alternating next-nearest-neighbor (NNN) and three-spin couplings in the presence of a uniform and a staggered magnetic field.

Keywords: Strongly correlated electrons; heavy fermions; quantized spin models.

PACS numbers: 71.27.+a, 75.10.Jm

1. Introduction

During the last decades, the correlation-induced metal-insulator (Mott) transition has been one of the challenging problems in condensed matter physics.¹ In most cases, the translational symmetry is broken in the Mott insulator.² A notable exception is the one-dimensional repulsive ($U > 0$) Hubbard model³

$$\mathcal{H}_{\text{Hub}} = t \sum_n \sum_{\alpha=\uparrow,\downarrow} (c_{n\alpha}^\dagger c_{n+1,\alpha} + c_{n+1,\alpha}^\dagger c_{n\alpha}) + U \sum_n \hat{\rho}_{n\uparrow} \hat{\rho}_{n\downarrow} \quad (1)$$

at half-filling, where the dynamical generation of a charge gap is not accompanied by the breaking of a discrete symmetry.⁴ In Eq. (1), we have used standard notation, namely $c_{n\alpha}^\dagger$ ($c_{n\alpha}$) for electron creation (annihilation) operators and $\hat{\rho}_{n\alpha} = c_{n\alpha}^\dagger c_{n\alpha}$ for the particle density at site n with spin projection α . The exact solution of the model (1) in the case of a half-filled band reveals that the ground state is uniform, with exponentially decaying density correlations.⁵ At the same time, spin excitations are gapless and thus magnetic correlations decay only algebraically.⁶ This is readily understood in the large- U limit: indeed, for $U \gg |t|$ the infrared behavior of the model (1) at half-filling is fully described by the SU(2)-symmetric spin $S = 1/2$ Heisenberg Hamiltonian

$$\mathcal{H}_{\text{Heis}} = J \sum_n \mathbf{S}_n \cdot \mathbf{S}_{n+1} + J' \sum_n \mathbf{S}_n \cdot \mathbf{S}_{n+2}, \quad (2)$$

where $J = 4t^2/U - 16t^4/U^3$ and $J' = 4t^4/U^3$ up to the fourth-order terms in t/U .^{7,8} Since the condition $|t| \ll U$ implies that the frustration is weak $J' \ll J$, the next-nearest exchange is irrelevant and the low-energy behavior of the initial electron system is governed by the standard isotropic Heisenberg Hamiltonian $\mathcal{H}_{\text{Heis}} = J \sum_n \mathbf{S}_n \cdot \mathbf{S}_{n+1}$. Elegant techniques have been developed for calculating higher order corrections to the Hamiltonian (2). These terms are also irrelevant and leave the featureless character of the ground state intact.⁹⁻¹¹

The spin sector may even remain translationally invariant in the case of an explicitly broken translational invariance of the electronic Hamiltonian. For example, let us consider a scenario where two types of atoms are located respectively on even and odd sites of the lattice, with different on-site energies and/or different on-site couplings for the electrons. The Hamiltonian of such an extended version of the Hubbard model is given by

$$\mathcal{H} = t \sum_{n,\alpha} (c_{n\alpha}^\dagger c_{n+1,\alpha} + c_{n+1,\alpha}^\dagger c_{n\alpha}) + U \sum_n (1 + (-1)^n \delta) \hat{\rho}_{n\uparrow} \hat{\rho}_{n\downarrow} + \frac{\Delta}{2} \sum_n \sum_\alpha (-1)^n \hat{\rho}_{n\alpha}, \quad (3)$$

where $0 \leq \delta, \Delta/U \ll 1$. It possesses spin SU(2) symmetry, but the translational symmetry has been reduced due to the doubling of the unit cell. At $\delta = 0$ and $\Delta \neq 0$ this Hamiltonian corresponds to the ionic Hubbard model (IHM),¹² where electrons on even and odd sites have different on-site energies $\pm\Delta/2$, while at $\Delta = 0$ and $\delta \neq 0$ Eq. (3) represents the alternating- U Hubbard model,¹³ where the electrons experience different on-site interactions on even and odd sites.

At $U = 0$, the half-filled IHM describes a regular band insulator with equal charge and spin gaps and a long-range ordered (LRO) charge density wave (CDW) in the ground state. With increasing U , the system undergoes two phase transitions, a first one at $U = U_{c1}$ from the CDW-insulator to a LRO dimerized insulator, and a second one at $U = U_{c2} > U_{c1}$ from the dimerized phase to a strongly correlated (Mott) insulator.¹⁴ At $U = U_{c2}$ the spin gap vanishes and the low-energy behavior of the system for $U > U_{c2}$ is again described by the Heisenberg Hamiltonian (2), with the difference that the spin exchange parameters J and J' now weakly depend on Δ .

The broken translational symmetry of the model manifests itself only in the charge degrees of freedom via the presence of a LRO CDW pattern which persists even in the limit of strong repulsion, with the amplitude approaching zero at $U \rightarrow \infty$.¹⁵

The weak-coupling renormalization group analysis of the repulsive alternating- U Hubbard model ($\Delta = 0$ and $U(1 \pm \delta) > 0$) shows a qualitatively similar low-energy behavior at half-filling as the usual Hubbard model. Scattering processes arising from the alternating part of the interaction, which are relevant in the commensurate case of 1/4- and 3/4-filled bands,¹³ are irrelevant at 1/2-filling where the properties of the system are governed by the uniform part of the interaction. In the limit of strong on-site repulsion ($U \gg |t|$), the infrared behavior of the alternating- U Hubbard model is once again described by the Heisenberg Hamiltonian (2), but with a slight modification — an alternating next-nearest-neighbor (NNN) exchange $\sum_n [J'_0 + (-1)^n J'_1] \mathbf{S}_n \cdot \mathbf{S}_{n+2}$.¹⁶ Numerical and analytical studies of the Heisenberg chain with alternating NNN exchange show that in the pertinent case of weak frustration ($J' \ll J$), the alternation of J' is irrelevant and the infrared behavior of the model is fully described by the standard Heisenberg model with nearest-neighbor exchange.¹⁷ Thus, even though the Hamiltonian (3) describes a fermion system on a lattice with broken translational symmetry, the information about the unit cell doubling at half-filling is fully accommodated within the high-energy degrees of freedom; the low-energy behavior of the system is described by a translationally invariant, isotropic spin Hamiltonian. It has to be noted that the above conclusion does not remain valid in the presence of bond alternation, i.e., if the hopping amplitude t is replaced by $t_0 + (-1)^n t_1$. In this case, one obtains that in the strong-coupling limit at half-filling the effective Hamiltonian, still given by the Heisenberg model, contains an alternating nearest-neighbor exchange $\sum_n [J_0 + (-1)^n J_1] \mathbf{S}_n \cdot \mathbf{S}_{n+1}$, which leads to the spin-Peierls instability with gapped spin excitations.¹⁸

We now turn our attention to a model having full translational symmetry, but explicitly broken spin $SU(2)$ symmetry, the so-called spin-asymmetric Hubbard Hamiltonian

$$\mathcal{H} = \sum_{n,\alpha} t_\alpha (c_{n\alpha}^\dagger c_{n+1,\alpha} + c_{n+1,\alpha}^\dagger c_{n\alpha}) + U \sum_n \hat{\rho}_{n\uparrow} \hat{\rho}_{n\downarrow}, \quad (4)$$

where the hopping is spin-dependent ($t_\uparrow \neq t_\downarrow$). This model, introduced in the early 1990s¹⁹ to interpolate between the standard Hubbard model ($t_\uparrow = t_\downarrow$) and the Falicov–Kimball model²⁰ ($t_\uparrow > 0$, $t_\downarrow = 0$), has been intensively studied during the last two decades.^{21–26} Away from half-filling the spin-up and spin-down particles are segregated in the ground state for large enough repulsion, both for the Falicov–Kimball model²⁷ and for the Hamiltonian (4) with $t_\uparrow \neq t_\downarrow \neq 0$.²⁸ Therefore, the spin-asymmetric Hubbard model appears to be well suited for studying transitions between phase-separated and homogeneous states, especially in one dimension.^{29–31}

More recently, increased interest in low dimensional correlated fermion models with spin-dependent hopping has been triggered by the fascinating progress in experimental studies of low dimensional mixtures of optically trapped ultracold atoms

of two different types,³² such as ultracold atoms loaded into spin-dependent optical lattices^{33,34} or trapped atoms of different masses.^{35,36} The great freedom available for generating optical lattices has also allowed one to play with the lattice geometry and to create bipartite lattices, which turned out to be a key ingredient for achieving higher-band condensates,^{37–39} coherence control,⁴⁰ density-wave dynamics⁴¹ and even graphene-like physics.^{42,43} It has to be emphasized that mixtures of fermions with different hopping amplitudes naturally appear in solid-state systems as well, namely when several bands cross the Fermi surface. This happens, for instance, in mixed-valence materials, organic superconductors,⁴⁴ small radius nanotubes⁴⁵ and even graphene-based heterostructures.⁴⁶ However, experiments with trapped ultracold atoms can actually engineer quantum many-body states and thus realize models of correlated fermions and bosons which are not available in usual solid-state structures.⁴⁷ Recent theoretical predictions of various unconventional superfluid or superconducting,^{48–54} insulating^{55,56} and magnetic⁵⁷ phases in such novel systems have further stimulated the interest in the spin-asymmetric Hubbard model.

The broken $SU(2)$ spin symmetry of the model (4) at $t_{\uparrow} \neq t_{\downarrow}$ is manifestly seen for a half-filled band in the strong-coupling limit ($U \gg |t_{\uparrow}|, |t_{\downarrow}|$), where to leading order the infrared behavior of the system is described by the anisotropic XXZ Heisenberg Hamiltonian

$$\mathcal{H} = J \sum_n (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y + \gamma S_n^z S_{n+1}^z), \quad (5)$$

with $J = 4t_{\uparrow}t_{\downarrow}/U$ and $\gamma = (t_{\uparrow}^2 + t_{\downarrow}^2)/2t_{\uparrow}t_{\downarrow}$.²² As the anisotropy parameter $|\gamma|$ is larger than 1 for arbitrary $t_{\uparrow} \neq t_{\downarrow}$, the system has a finite spin gap and long-range antiferromagnetic order in the ground state.⁵⁸ Nevertheless, the translational invariance of the initial lattice model (4) is retained by the effective Hamiltonian (5), even if the ground state has lower symmetry due to the general phenomenon of spontaneous symmetry breaking.

In a recent paper, we have studied the one-dimensional spin-asymmetric IHM in the limit of strong on-site repulsion (for a half-filled band).⁵⁹ We have shown that for $t_{\uparrow} \neq t_{\downarrow}$ the doubling of the unit cell by the alternating ionic potential $\Delta \neq 0$ directly manifests itself in the spin degrees of freedom, and the effective spin Hamiltonian in the strong-coupling limit is given by the anisotropic XXZ Heisenberg chain with a staggered magnetic field

$$\mathcal{H} = J \sum_n (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y + \gamma S_n^z S_{n+1}^z) - h \sum_n (-1)^n S_n^z, \quad (6)$$

where

$$J = \frac{4t_{\uparrow}t_{\downarrow}}{U(1-x^2)}, \quad \gamma = \frac{t_{\uparrow}^2 + t_{\downarrow}^2}{2t_{\uparrow}t_{\downarrow}}, \quad h = \frac{2(t_{\uparrow}^2 - t_{\downarrow}^2)x}{U(1-x^2)}, \quad (7)$$

and $x = \Delta/U$. For $t_{\uparrow} \neq t_{\downarrow}$ and finite x , the translational symmetry is broken already at the level of the effective spin Hamiltonian via the presence of the staggered

magnetic field. Since this represents a strongly relevant perturbation to the spin system, the ground state is characterized by a long-range antiferromagnetic order with explicitly broken translational symmetry. The excitation spectrum is gapped and the gap exhibits power-law dependence on the parameter h .⁶⁰

In the present paper, we extend our analysis to the case of explicitly broken translational symmetry in the on-site interaction and derive the effective spin Hamiltonian for the one-dimensional spin-asymmetric alternating- U ionic Hubbard chain represented by

$$\mathcal{H} = \sum_{n,\alpha} t_\alpha (c_{n\alpha}^\dagger c_{n+1,\alpha} + c_{n+1,\alpha}^\dagger c_{n\alpha}) + U \sum_n (1 + (-1)^n \delta) \hat{\rho}_{n\uparrow} \hat{\rho}_{n\downarrow} + \frac{\Delta}{2} \sum_n \sum_\alpha (-1)^n \hat{\rho}_{n\alpha}. \quad (8)$$

We find that up to fourth-order terms in t_α/U , the infrared behavior of the lattice fermion model (8) at half-filling is governed by the following effective spin Hamiltonian:

$$\begin{aligned} \mathcal{H}_{\text{eff}} = \sum_n \bigg\{ & J_\perp (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) + J_\parallel \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) \\ & + J'_\perp(n) (S_n^x S_{n+2}^x + S_n^y S_{n+2}^y) + J'_\parallel(n) \left(S_n^z S_{n+2}^z - \frac{1}{4} \right) \\ & + W_\perp(n) [(S_{n-1}^x S_n^x + S_{n-1}^y S_n^y) S_{n+1}^z + S_{n-1}^z (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y)] \\ & + W_\parallel(n) S_{n-1}^z S_n^z S_{n+1}^z - h(n) S_n^z \bigg\}, \quad (9) \end{aligned}$$

where

$$J_\perp = \frac{4t_\uparrow t_\downarrow}{U(1-\lambda^2)} \left[1 - \frac{2(t_\uparrow^2 + t_\downarrow^2)}{U^2(1-\lambda^2)^2} \left(2 + 2\lambda^2 - \frac{1-\lambda^2}{1-\delta^2} \right) \right], \quad (10)$$

$$J_\parallel = \frac{2(t_\uparrow^2 + t_\downarrow^2)}{U(1-\lambda^2)} - \frac{6(t_\uparrow^4 + t_\downarrow^4)}{U^3(1-\lambda^2)^3} (1 + 3\lambda^2) - \frac{4t_\uparrow^2 t_\downarrow^2}{U^3(1-\lambda^2)^3} \left(5 - \lambda^2 - \frac{4(1-\lambda^2)}{1-\delta^2} \right), \quad (11)$$

$$J'_\perp(n) = \frac{4t_\uparrow^2 t_\downarrow^2}{U^3(1-\lambda^2)^3} \left[2 + 2\lambda^2 - \frac{(1-\lambda^2)^2}{1-\delta^2} \right] + (-1)^n \frac{4\delta t_\uparrow^2 t_\downarrow^2}{U^3(1-\lambda^2)(1-\delta^2)}, \quad (12)$$

$$\begin{aligned} J'_\parallel(n) = & \frac{2(t_\uparrow^4 + t_\downarrow^4)}{U^3(1-\lambda^2)^3} \left[1 + 3\lambda^2 + \frac{1-\lambda^4}{1-\delta^2} \right] - \frac{4t_\uparrow^2 t_\downarrow^2}{U^3(1-\lambda^2)^2} \frac{1+\delta^2}{1-\delta^2} \\ & + (-1)^n \frac{2\delta}{U^3(1-\lambda^2)^2(1-\delta^2)} [4t_\uparrow^2 t_\downarrow^2 - (t_\uparrow^4 + t_\downarrow^4)(1+\lambda^2)], \quad (13) \end{aligned}$$

$$W_\perp(n) = \frac{4\lambda t_\uparrow t_\downarrow (t_\uparrow^2 - t_\downarrow^2)}{U^3(1-\lambda^2)^2} \left\{ \frac{2\delta}{1-\delta^2} + (-1)^n \left[\frac{3+\lambda^2}{1-\lambda^2} + \frac{2}{1-\delta^2} \right] \right\}, \quad (14)$$

$$W_{\parallel}(n) = \frac{4\lambda(t_{\uparrow}^4 - t_{\downarrow}^4)}{U^3(1 - \lambda^2)^2} \left\{ \frac{2\delta}{1 - \delta^2} + (-1)^n \left[\frac{3 + \lambda^2}{1 - \lambda^2} + \frac{2}{1 - \delta^2} \right] \right\}, \quad (15)$$

$$h(n) = h_0 + (-1)^n h_1 = \frac{2\lambda\delta(t_{\uparrow}^4 - t_{\downarrow}^4)}{U^3(1 - \lambda^2)^2(1 - \delta^2)} + (-1)^n \frac{2\lambda(t_{\uparrow}^2 - t_{\downarrow}^2)}{U(1 - \lambda^2)} \left\{ 1 - \frac{t_{\uparrow}^2 + t_{\downarrow}^2}{2U^2(1 - \lambda^2)^2} \left[5(3 + \lambda^2) - \frac{2(1 - \lambda^2)}{1 - \delta^2} \right] \right\}, \quad (16)$$

with $\lambda = \delta + \Delta/U$.

As we observe, the leading terms ($\propto U^{-1}$) are the same as in (7) except that the parameter x is replaced by λ . The higher order terms ($\propto U^{-3}$) include the renormalization of the nearest-neighbor coupling J , the NNN exchange with an alternating part whose existence is determined solely by δ , and corrections to the magnetic field alongside the less conventional three-spin terms, all having both homogeneous and alternating parts. We also note that the expressions of the fourth-order terms obtained in our earlier work⁵⁹ are not entirely correct and they should be replaced by the appropriate limit ($\delta = 0$) of the above results.

A detailed derivation of the expressions (9)–(16) will be presented in the following. In Secs. 2 and 3, a unitary transformation is applied to the electronic Hamiltonian in the case of a half-filled band, eliminating hopping processes between many-electron states with different numbers of doubly occupied sites. In Sec. 4 we briefly discuss the Hubbard operators, which are used in the subsequent Sec. 5 to derive the effective spin Hamiltonian. Finally, Sec. 6 summarizes the main results of the paper, while the Appendix contains some technical calculations of the spin exchange terms.

2. The Strong-coupling Approach

In the strong coupling limit ($U \gg |t|$), the perturbative treatment of the half-filled Hubbard model based on expansion of the Hamiltonian in powers of t/U goes back to the original derivation of the effective spin Hamiltonian to the second order by Anderson.⁶¹ Afterwards, using different versions of the degenerate perturbation theory, effective spin Hamiltonians up to higher orders in t/U have been obtained. In particular, Klein and Seitz⁶² derived the sixth-order spin interaction for the Hubbard chain, while Bulaevskii⁷ and Takahashi⁸ obtained the fourth-order terms for the half-filled Hubbard model in higher dimensions. More recently, these perturbative methods have also been applied to Hubbard models with more general interactions.⁶³

An alternative approach to construct the effective Hamiltonian is based on unitary transformations. Harris and Lange⁶⁴ used such a transformation to obtain second-order results and to calculate spectral properties of the Hubbard model. A transformation which systematically incorporates higher orders in t/U has been proposed by Chao *et al.*⁶⁵ In their expansion, closed expressions for the effective

spin exchange are obtained to any order. However, beyond the second order their method is not very well controlled since the transformation of the Hamiltonian involves an approximation for the band energies, and higher order terms mixing different Hubbard bands are not eliminated properly.^{66,67}

A consistent transformation scheme which allows one to remove all unphysical terms and to derive the t/U -expansion up to any desired order has been formulated by MacDonald *et al.*⁹ In their scheme, interaction terms which do not conserve the number of local electron pairs are eliminated from the Hamiltonian order by order in an iterative treatment, generating new interactions and thus improving the accuracy of the transformation at each step. Later their approach has been successfully employed to obtain effective spin Hamiltonians in the case of extended versions of the Hubbard model on a square lattice with next-nearest- and next-NNN hoppings.^{68,69}

Another consistent scheme for construction of the effective spin Hamiltonian up to any given order in powers of t/U has been developed by Stein,¹⁰ who utilized Wegner's method⁷⁰ of continuous unitary transformations with subsequent solution of the corresponding flow equations for the half-filled Hubbard model. Later a similar approach has been used to reveal an additional (hidden) symmetry of the Hubbard model on any bipartite lattice.⁷¹

In this paper we apply the method developed by MacDonald *et al.* for the standard Hubbard model⁹ to the Hamiltonian $\mathcal{H} = T + V$, where

$$T = \sum_{\langle n,m \rangle} \sum_{\alpha} t_{\alpha} c_{n\alpha}^{\dagger} c_{m\alpha}, \quad (17)$$

$$V = \frac{\Delta}{2} \sum_{n,\alpha} (-1)^n \hat{\rho}_{n\alpha} + U_o \sum_n \hat{\rho}_{2n+1,\uparrow} \hat{\rho}_{2n+1,\downarrow} + U_e \sum_n \hat{\rho}_{2n,\uparrow} \hat{\rho}_{2n,\downarrow}, \quad (18)$$

and brackets in the sum $\langle n, m \rangle$ signify that n and m are labels for neighboring sites. The on-site couplings $U_o = U(1 - \delta)$ and $U_e = U(1 + \delta)$ are supposed to be strong, $U_e \geq U_o \gg |t_{\uparrow}|, |t_{\downarrow}|, \Delta$, implying that the parameters $\delta = (U_e - U_o)/(U_e + U_o)$ and $\lambda = \delta + \Delta/U$ satisfy the conditions $0 \leq \delta < 1$, $0 \leq \lambda < 1$.

In the large- U limit of the standard Hubbard model ($\delta = \Delta = 0$) the many-electron states are grouped according to the number of doubly occupied sites (doublets) N_d . In the present case with $\delta, \Delta > 0$ these Hubbard subbands are split into groups of states classified by two numbers, N_{de} and N_{do} , representing the numbers of doubly occupied sites on even and odd sublattices, respectively. The hopping operator T mixes the states of these subbands. The “unmixing” can be achieved by introducing suitable linear combinations of the uncorrelated basis states. The \mathcal{S} -matrix for this transformation, and the transformed Hamiltonian,

$$\mathcal{H}' = e^{i\mathcal{S}} \mathcal{H} e^{-i\mathcal{S}}, \quad (19)$$

are generated by an iterative procedure, which results in an expansion in powers of the hopping amplitudes t_{\uparrow} and/or t_{\downarrow} divided by the on-site energies U_e and/or U_o .

This expansion is based on the separation of the kinetic part of the Hamiltonian into three terms:

$$T = T_0 + T_1 + T_{-1}, \quad (20)$$

where T_0 leaves the number of doubly occupied sites unchanged, and T_1 (T_{-1}) increases (decreases) this number by one. In the present case of broken translational symmetry each of these contributions is further split into several different terms, depending on whether the electron hops from an even to an odd site or vice versa.

In particular, the T_0 term is split into four separate processes:

$$T_0 = T_0^{\text{pe}} + T_0^{\text{po}} + T_0^{\text{de}} + T_0^{\text{do}}. \quad (21)$$

Here,

$$T_0^{\text{pe}} = \sum_{\langle 2n, m \rangle} \sum_{\alpha} t_{\alpha} (1 - \hat{\rho}_{2n, \bar{\alpha}}) c_{2n, \alpha}^{\dagger} c_{m\alpha} (1 - \hat{\rho}_{m\bar{\alpha}}) \quad (22)$$

and

$$T_0^{\text{po}} = \sum_{\langle 2n+1, m \rangle} \sum_{\alpha} t_{\alpha} (1 - \hat{\rho}_{2n+1, \bar{\alpha}}) c_{2n+1, \alpha}^{\dagger} c_{m\alpha} (1 - \hat{\rho}_{m\bar{\alpha}}) \quad (23)$$

correspond respectively to hopping processes where an electron with spin α hops from a singly occupied odd (even) site to an empty neighboring even (odd) site, while

$$T_0^{\text{de}} = \sum_{\langle 2n, m \rangle} \sum_{\alpha} t_{\alpha} \hat{\rho}_{2n, \bar{\alpha}} c_{2n, \alpha}^{\dagger} c_{m\alpha} \hat{\rho}_{m\bar{\alpha}} \quad (24)$$

and

$$T_0^{\text{do}} = \sum_{\langle 2n+1, m \rangle} \sum_{\alpha} t_{\alpha} \hat{\rho}_{2n+1, \bar{\alpha}} c_{2n+1, \alpha}^{\dagger} c_{m\alpha} \hat{\rho}_{m\bar{\alpha}} \quad (25)$$

represent hopping processes where an electron with spin α hops from a doubly occupied odd (even) site to a neighboring even (odd) site which is already occupied by another electron with the opposite spin $\bar{\alpha}$.

In a similar fashion, the operators $T_{\pm 1}$, which change the number of doublons by one, are also separated into even and odd parts $T_{\pm 1} = T_{\pm 1}^{\text{e}} + T_{\pm 1}^{\text{o}}$, where

$$T_1^{\text{e}} = \sum_{\langle 2n, m \rangle} \sum_{\alpha} t_{\alpha} \hat{\rho}_{2n, \bar{\alpha}} c_{2n, \alpha}^{\dagger} c_{m\alpha} (1 - \hat{\rho}_{m\bar{\alpha}}) \quad (26)$$

and

$$T_1^{\text{o}} = \sum_{\langle 2n+1, m \rangle} \sum_{\alpha} t_{\alpha} \hat{\rho}_{2n+1, \bar{\alpha}} c_{2n+1, \alpha}^{\dagger} c_{m\alpha} (1 - \hat{\rho}_{m\bar{\alpha}}) \quad (27)$$

increase the number of doublons on the sublattice of even (odd) sites, while

$$T_{-1}^{\text{e}} = \sum_{\langle n, 2m \rangle} \sum_{\alpha} t_{\alpha} (1 - \hat{\rho}_{n\bar{\alpha}}) c_{n\alpha}^{\dagger} c_{2m, \alpha} \hat{\rho}_{2m, \bar{\alpha}} \quad (28)$$

and

$$T_{-1}^o = \sum_{\langle n, 2m+1 \rangle} \sum_{\alpha} t_{\alpha} (1 - \hat{\rho}_{n\bar{\alpha}}) c_{n\alpha}^{\dagger} c_{2m+1, \alpha} \hat{\rho}_{2m+1, \bar{\alpha}}, \quad (29)$$

respectively decrease the number of doublons on the even and odd sublattices.

One can easily check the following commutation relations:

$$[V, T_{\mu}^s] = (\mu + \delta_{\mu,0}) \Lambda_s T_{\mu}^s, \quad (30)$$

where $\mu = 0, \pm 1$ and

$$\Lambda_s = \begin{cases} \Delta, & s = \text{pe}, \\ -\Delta, & s = \text{po}, \\ (U_e - U_o) + \Delta, & s = \text{de}, \\ -(U_e - U_o) - \Delta, & s = \text{do}, \\ U_e + \Delta, & s = \text{e}, \\ U_o - \Delta, & s = \text{o}. \end{cases} \quad (31)$$

The relations (30) reflect the fact that the energy of the system changes by $(\mu + \delta_{\mu,0}) \Lambda_s$ as a result of the hopping process T_{μ}^s .

3. Effective Hamiltonian in the Case of a Half-filled Band

Let us now search for the unitary transformation \mathcal{S} which eliminates hops between states with different numbers of doubly occupied sites in the transformed Hamiltonian

$$\mathcal{H}' = e^{i\mathcal{S}} \mathcal{H} e^{-i\mathcal{S}} = \mathcal{H} + [i\mathcal{S}, \mathcal{H}] + \frac{1}{2}[i\mathcal{S}, [i\mathcal{S}, \mathcal{H}]] + \dots \quad (32)$$

We follow a recursive scheme⁹ which allows to determine such a transformation to any desired order in t_{α}/U . The last two terms of the initial Hamiltonian

$$\mathcal{H} \equiv \mathcal{H}'^{(1)} = V + T_0 + T_1 + T_{-1} \quad (33)$$

may be transformed away by choosing

$$i\mathcal{S} \equiv i\mathcal{S}^{(1)} = \frac{1}{U_o - \Delta} (T_1^o - T_{-1}^o) + \frac{1}{U_e + \Delta} (T_1^e - T_{-1}^e). \quad (34)$$

Substituting (33) and (34) into the expansion (32) and applying (30), we obtain

$$\begin{aligned} \mathcal{H}'^{(2)} = & V + T_0 + \frac{1}{U_o - \Delta} ([T_1^o, T_0] + [T_0, T_{-1}^o] + [T_1^o, T_{-1}^o]) \\ & + \frac{1}{U_e + \Delta} ([T_1^e, T_0] + [T_0, T_{-1}^e] + [T_1^e, T_{-1}^e]) \\ & + \frac{U_e + U_o}{2(U_e + \Delta)(U_o - \Delta)} ([T_1^e, T_{-1}^o] + [T_1^o, T_{-1}^e]) \\ & + \frac{U_o - U_e - 2\Delta}{2(U_e + \Delta)(U_o - \Delta)} ([T_1^e, T_1^o] + [T_{-1}^o, T_{-1}^e]) + \mathcal{O}(t^3/U^2). \end{aligned} \quad (35)$$

We focus on the case of a half-filled band, where in the large- U limit the lowest-energy states $|\Psi_L\rangle$ have exactly one electron at each site. In this subspace no hops are possible without increasing the number of doubly occupied sites. Therefore,

$$T_{-1}^e |\Psi_L\rangle = 0, \quad T_{-1}^o |\Psi_L\rangle = 0, \quad T_0 |\Psi_L\rangle = 0 \quad (36)$$

and the effective Hamiltonian (35) is reduced to

$$\mathcal{H}'^{(2)} = -\frac{T_{-1}^o T_1^o}{U_o - \Delta} - \frac{T_{-1}^e T_1^e}{U_e + \Delta} + \mathcal{O}(t^3/U^2). \quad (37)$$

To proceed further, we define:

$$T^{(k)}[\{s\}, \{\mu\}] = T_{\mu_1}^{s_1} T_{\mu_2}^{s_2} \dots T_{\mu_k}^{s_k}. \quad (38)$$

Using (30), we can write

$$[\hat{V}, T^{(k)}[\{s\}, \{\mu\}]] = \sum_{i=1}^k \Lambda_{s_i} (\mu_i + \delta_{\mu_i, 0}) T^{(k)}[\{s\}, \{\mu\}]. \quad (39)$$

$\mathcal{H}'^{(k)}$ contains terms of order $(t_\alpha)^k$, denoted by $\mathcal{H}'^{[k]}$, which couple states in different subspaces. By definition $[V, \mathcal{H}'^{[k]}] \neq 0$ and $\mathcal{H}'^{[k]}$ can be expressed in the following way:

$$\mathcal{H}'^{[k]} = \sum_{\{a\}} \sum_{\{\mu\}} C_{\{a\}}^{(k)}(\{\mu\}) T^{(k)}[\{a\}, \{\mu\}], \quad \sum_{i=1}^k \mu_i \neq 0. \quad (40)$$

If at each k th order step we choose $\mathcal{S}^{(k)}$ as

$$\mathcal{S}^{(k)} = \mathcal{S}^{(k-1)} + \mathcal{S}^{[k]}, \quad (41)$$

where $\mathcal{S}^{[k]}$ is the solution of the equation

$$[i\mathcal{S}^{[k]}, V] = -\mathcal{H}'^{[k]} \quad (42)$$

and therefore equals

$$\mathcal{S}^{[k]} = -i \sum_{\{a\}} \sum_{\{\mu\}} \frac{C_{\{a\}}^{(k)}(\{\mu\})}{\sum_{i=1}^k \Lambda_{a_i} (\mu_i + \delta_{\mu_i, 0})} T^{(k)}[\{a\}, \{\mu\}], \quad \sum_{i=1}^k \mu_i \neq 0, \quad (43)$$

then the transformed Hamiltonian

$$\mathcal{H}'^{(k+1)} = e^{i\mathcal{S}^{(k)}} \mathcal{H} e^{-i\mathcal{S}^{(k)}} \quad (44)$$

contains terms up to the order of $(t_\alpha)^k/U^{k-1}$ which commute with the unperturbed Hamiltonian V and mix states within each subspace only.

The conditions (36) can be generalized to higher orders

$$T^{(k)}[\{s\}, \{\mu\}] |\Psi_L\rangle = 0, \quad (45)$$

if

$$M_p^k[\{\mu\}] \equiv \sum_{i=p}^k \mu_i < 0 \quad (46)$$

for at least one value of p . Equation (45) can be used to eliminate many terms from the expansion for \mathcal{H}' in the subspace of minimal $\langle V \rangle$.

The final expression of the transformed Hamiltonian \mathcal{H}' up to the fourth order reads:

$$\begin{aligned}
\mathcal{H}^{(4)} = & -\frac{T_{-1}^o T_1^o}{U_o - \Delta} - \frac{T_{-1}^e T_1^e}{U_e + \Delta} - \frac{T_{-1}^o T_0^{po} T_0^{pe} T_1^o}{U_o(U_o - \Delta)^2} - \frac{T_{-1}^o T_0^{do} T_0^{de} T_1^o}{U_e(U_o - \Delta)^2} - \frac{T_{-1}^e T_0^{de} T_0^{do} T_1^e}{U_o(U_e + \Delta)^2} \\
& - \frac{T_{-1}^e T_0^{pe} T_0^{po} T_1^e}{U_e(U_e + \Delta)^2} - \frac{T_{-1}^o T_0^{po} T_0^{do} T_1^e}{(U_o - \Delta)U_o(U_e + \Delta)} - \frac{T_{-1}^e T_0^{de} T_0^{pe} T_1^o}{(U_o - \Delta)U_o(U_e + \Delta)} \\
& - \frac{T_{-1}^o T_0^{do} T_0^{po} T_1^e}{(U_o - \Delta)U_e(U_e + \Delta)} - \frac{T_{-1}^e T_0^{pe} T_0^{de} T_1^o}{(U_o - \Delta)U_e(U_e + \Delta)} \\
& - \frac{T_{-1}^o T_{-1}^e T_1^o T_1^e}{(U_e + \Delta)(U_o - \Delta)(U_e + U_o)} - \frac{T_{-1}^e T_{-1}^o T_1^e T_1^o}{(U_e + \Delta)(U_o - \Delta)(U_e + U_o)} \\
& - \frac{T_{-1}^e T_{-1}^o T_1^e T_1^e}{(U_e + \Delta)^2(U_e + U_o)} - \frac{T_{-1}^o T_{-1}^e T_1^o T_1^o}{(U_o - \Delta)^2(U_e + U_o)} \\
& - \frac{T_{-1}^o T_{-1}^o T_1^o T_1^o}{2(U_o - \Delta)^3} - \frac{T_{-1}^e T_{-1}^e T_1^e T_1^e}{2(U_e + \Delta)^3} + \frac{T_{-1}^o T_1^o T_{-1}^o T_1^o}{(U_o - \Delta)^3} + \frac{T_{-1}^e T_1^e T_{-1}^e T_1^e}{(U_e + \Delta)^3} \\
& + \frac{U_o + U_e}{2(U_o - \Delta)^2(U_e + \Delta)^2} [T_{-1}^o T_1^o T_{-1}^e T_1^e + T_{-1}^e T_1^e T_{-1}^o T_1^o]. \tag{47}
\end{aligned}$$

4. Hubbard Operators

To handle the effects of strong interaction properly, it is important to know whether at the beginning or at the end of a given hopping process a particular site is doubly occupied or not. For this purpose, one introduces the so-called Hubbard operators⁷² X_n^{ab} , which are defined at each site of the lattice and describe all possible transitions between the local basis states $|a\rangle$, $|b\rangle$: unoccupied $|0\rangle$, singly occupied with an up-spin $|\uparrow\rangle$ or a down-spin $|\downarrow\rangle$ electron, and doubly occupied $|2\rangle$. The original electron creation (annihilation) operators can be expressed in terms of Hubbard operators in the following way:

$$c_{n,\alpha}^\dagger = X_n^{\alpha 0} + \eta(\alpha) X_n^{2\bar{\alpha}}, \quad c_{n,\alpha} = X_n^{0\alpha} + \eta(\alpha) X_n^{\bar{\alpha} 2}, \tag{48}$$

where $\eta(\alpha) = \begin{cases} 1 & \text{if } \alpha = \uparrow, \\ -1 & \text{if } \alpha = \downarrow. \end{cases}$

Conversely, in terms of creation (annihilation) operators the Hubbard operators have the form:

$$\begin{aligned}
X_n^{\alpha 0} &= c_{n\alpha}^\dagger (1 - \hat{\rho}_{n\bar{\alpha}}), & X_n^{2\alpha} &= \eta(\bar{\alpha}) c_{i\bar{\alpha}}^\dagger \hat{\rho}_{i\alpha}, \\
X_n^{\alpha \bar{\alpha}} &= c_{n\alpha}^\dagger c_{n\bar{\alpha}}, & X_n^{20} &= \eta(\bar{\alpha}) c_{n\bar{\alpha}}^\dagger c_{n\alpha}^\dagger, \\
X_n^{00} &= (1 - \hat{\rho}_{n\uparrow})(1 - \hat{\rho}_{n\downarrow}), & X_n^{22} &= \hat{\rho}_{n\uparrow} \hat{\rho}_{n\downarrow}, \\
X_n^{\alpha \alpha} &= \hat{\rho}_{n\alpha} (1 - \hat{\rho}_{n\bar{\alpha}}).
\end{aligned} \tag{49}$$

The Hubbard operators containing an even (odd) number of electron creation/annihilation operators are Bose-like (Fermi-like) operators. They obey the following on-site multiplication rule:

$$X_n^{pq} X_n^{rs} = \delta_{qr} X_n^{ps} \quad (50)$$

and commutation relations

$$[X_n^{pq}, X_m^{rs}]_{\pm} = \delta_{nm} (\delta_{qr} X_m^{ps} \pm \delta_{ps} X_m^{rq}), \quad (51)$$

where the upper sign stands for the case when both operators are Fermi-like, otherwise the lower sign should be adopted.

It is straightforward to represent the hopping terms introduced in Sec. 2 by the Hubbard operators:

$$\begin{aligned} T_0^{\text{po}} &= \sum_{\langle 2n+1, m \rangle} \sum_{\alpha} t_{\alpha} X_{2n+1}^{\alpha 0} X_m^{0\alpha}, & T_0^{\text{pe}} &= \sum_{\langle 2n, m \rangle} \sum_{\alpha} t_{\alpha} X_{2n}^{\alpha 0} X_m^{0\alpha}, \\ T_0^{\text{do}} &= \sum_{\langle 2n+1, m \rangle} \sum_{\alpha} t_{\alpha} X_{2n+1}^{2\bar{\alpha}} X_m^{\bar{\alpha} 2}, & T_0^{\text{de}} &= \sum_{\langle 2n, m \rangle} \sum_{\alpha} t_{\alpha} X_{2n}^{2\bar{\alpha}} X_m^{\bar{\alpha} 2}, \\ T_1^{\text{o}} &= \sum_{\langle 2n+1, m \rangle} \sum_{\alpha} \eta(\alpha) t_{\alpha} X_{2n+1}^{2\bar{\alpha}} X_m^{0\alpha}, & T_1^{\text{e}} &= \sum_{\langle 2n, m \rangle} \sum_{\alpha} \eta(\alpha) t_{\alpha} X_{2n}^{2\bar{\alpha}} X_m^{0\alpha}, \\ T_{-1}^{\text{o}} &= \sum_{\langle n, 2m+1 \rangle} \sum_{\alpha} \eta(\alpha) t_{\alpha} X_n^{\alpha 0} X_{2m+1}^{\bar{\alpha} 2}, & T_{-1}^{\text{e}} &= \sum_{\langle n, 2m \rangle} \sum_{\alpha} \eta(\alpha) t_{\alpha} X_n^{\alpha 0} X_{2m}^{\bar{\alpha} 2}. \end{aligned} \quad (52)$$

One also easily verifies that the X -operators describing the transitions between singly occupied states can be rewritten in terms of spin $S = 1/2$ operators as

$$\begin{aligned} X_n^{\uparrow\downarrow} &= c_{n\uparrow}^{\dagger} c_{n\downarrow} = S_n^+, & X_n^{\downarrow\uparrow} &= c_{n\downarrow}^{\dagger} c_{n\uparrow} = S_n^-, \\ X_n^{\uparrow\uparrow} &= \frac{1}{2} + S_n^z, & X_n^{\downarrow\downarrow} &= \frac{1}{2} - S_n^z. \end{aligned} \quad (53)$$

5. The Spin Hamiltonian

Using the relations (52) and (53), it is straightforward to rewrite the products of T -terms in (47) via the Hubbard X and hence the spin $S = 1/2$ operators. We first consider the simplest two-component T -terms at great length to elucidate the procedure for more complicated contributions.

5.1. The second-order terms

Let us start from the hopping term which corresponds to creation and subsequent annihilation of a single doublon on an even $2n$ th site. Since the electron hopping is restricted to nearest-neighbor sites, this process only includes electrons located on two neighboring sites $2n \pm 1$, and is given by:

$$T_{-1}^{\text{e}} T_1^{\text{e}} = \sum_{n=1}^{L/2} \sum_{q=\pm 1} \sum_{\alpha} [t_{\alpha}^2 X_{2n+q}^{\alpha 0} X_{2n}^{\bar{\alpha} 2} X_{2n}^{2\bar{\alpha}} X_{2n+q}^{0\alpha} - t_{\alpha} t_{\bar{\alpha}} X_{2n+q}^{\alpha 0} X_{2n}^{\bar{\alpha} 2} X_{2n}^{2\alpha} X_{2n+q}^{0\bar{\alpha}}]$$

$$\begin{aligned}
&= \sum_{n=1}^{L/2} \sum_{q=\pm 1} \sum_{\alpha} [t_{\alpha}^2 X_{2n+q}^{\alpha\alpha} X_{2n}^{\bar{\alpha}\bar{\alpha}} - t_{\alpha} t_{\bar{\alpha}} X_{2n+q}^{\alpha\bar{\alpha}} X_{2n}^{\bar{\alpha}\alpha}] \\
&= \sum_{n=1}^{L/2} \sum_{q=\pm 1} [t_{\uparrow}^2 X_{2n+q}^{\uparrow\uparrow} X_{2n}^{\downarrow\downarrow} + t_{\downarrow}^2 X_{2n+q}^{\downarrow\downarrow} X_{2n}^{\uparrow\uparrow} - t_{\uparrow} t_{\downarrow} (X_{2n+q}^{\uparrow\downarrow} X_{2n}^{\downarrow\uparrow} + X_{2n+q}^{\downarrow\uparrow} X_{2n}^{\uparrow\downarrow})] \\
&= \sum_{n=1}^{L/2} \sum_{q=\pm 1} \left[t_{\uparrow}^2 \left(\frac{1}{2} + S_{2n+q}^z \right) \left(\frac{1}{2} - S_{2n}^z \right) + t_{\downarrow}^2 \left(\frac{1}{2} - S_{2n+q}^z \right) \left(\frac{1}{2} + S_{2n}^z \right) \right. \\
&\quad \left. - t_{\uparrow} t_{\downarrow} (S_{2n+q}^+ S_{2n}^- + S_{2n+q}^- S_{2n}^+) \right] \\
&= \sum_{n=1}^{L/2} \sum_{q=\pm 1} \left[(t_{\uparrow}^2 + t_{\downarrow}^2) \left(\frac{1}{4} - S_{2n}^z S_{2n+q}^z \right) - \frac{1}{2} (t_{\uparrow}^2 - t_{\downarrow}^2) (S_{2n}^z - S_{2n+q}^z) \right. \\
&\quad \left. - 2t_{\uparrow} t_{\downarrow} (S_{2n}^x S_{2n+q}^x + S_{2n}^y S_{2n+q}^y) \right] \\
&= \sum_{n=1}^L \left[-2t_{\uparrow} t_{\downarrow} (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) - (t_{\uparrow}^2 + t_{\downarrow}^2) \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) \right. \\
&\quad \left. - (-1)^n (t_{\uparrow}^2 - t_{\downarrow}^2) S_n^z \right]. \tag{54}
\end{aligned}$$

As to the second term $T_{-1}^{\circ} T_1^{\circ}$, which describes creation and annihilation of a pair on an odd site, the calculation is essentially the same, with the only difference being that one needs to make a replacement $2n \leftrightarrow 2n+1$. Below this shift is absorbed in q :

$$\begin{aligned}
T_{-1}^{\circ} T_1^{\circ} &= \sum_{n=1}^{L/2} \sum_{q=0,2} \sum_{\alpha} [t_{\alpha}^2 X_{2n+q}^{\alpha 0} X_{2n+1}^{\bar{\alpha} 2} X_{2n+1}^{2\bar{\alpha}} X_{2n+q}^{0\alpha} \\
&\quad - t_{\alpha} t_{\bar{\alpha}} X_{2n+q}^{\alpha 0} X_{2n+1}^{\bar{\alpha} 2} X_{2n+1}^{2\alpha} X_{2n+q}^{0\bar{\alpha}}] \\
&= \sum_{n=1}^{L/2} \sum_{q=0,2} \left[t_{\uparrow}^2 \left(\frac{1}{2} + S_{2n+q}^z \right) \left(\frac{1}{2} - S_{2n+1}^z \right) + t_{\downarrow}^2 \left(\frac{1}{2} - S_{2n+q}^z \right) \left(\frac{1}{2} + S_{2n+1}^z \right) \right. \\
&\quad \left. - t_{\uparrow} t_{\downarrow} (S_{2n+q}^+ S_{2n+1}^- + S_{2n+q}^- S_{2n+1}^+) \right] \\
&= \sum_{n=1}^{L/2} \sum_{q=0,2} \left[(t_{\uparrow}^2 + t_{\downarrow}^2) \left(\frac{1}{4} - S_{2n+q}^z S_{2n+1}^z \right) + \frac{1}{2} (t_{\uparrow}^2 - t_{\downarrow}^2) (S_{2n+q}^z - S_{2n+1}^z) \right. \\
&\quad \left. - 2t_{\uparrow} t_{\downarrow} (S_{2n+q}^x S_{2n+1}^x + S_{2n+q}^y S_{2n+1}^y) \right]
\end{aligned}$$

$$= \sum_{n=1}^L \left[-2t_{\uparrow}t_{\downarrow}(S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) - (t_{\uparrow}^2 + t_{\downarrow}^2) \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) + (-1)^n (t_{\uparrow}^2 - t_{\downarrow}^2) S_n^z \right]. \quad (55)$$

The combination of these two processes yields the second-order effective spin Hamiltonian:

$$\begin{aligned} \mathcal{H}_{\text{eff}}^{(2)} &= -\frac{1}{U_o - \Delta} T_{-1}^o T_1^o - \frac{1}{U_e + \Delta} T_{-1}^e T_1^e \\ &= J \sum_n \left[S_n^x S_{n+1}^x + S_n^y S_{n+1}^y + \gamma \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) \right] - h \sum_n (-1)^n S_n^z, \end{aligned} \quad (56)$$

where

$$J = \frac{4t_{\uparrow}t_{\downarrow}}{U(1-\lambda^2)}, \quad \gamma = \frac{t_{\uparrow}^2 + t_{\downarrow}^2}{2t_{\uparrow}t_{\downarrow}}, \quad h = \frac{2\lambda(t_{\uparrow}^2 - t_{\downarrow}^2)}{U(1-\lambda^2)}. \quad (57)$$

As we see, the second-order effective Hamiltonian, which describes the spin degrees of freedom of the initial lattice fermion model, is the Hamiltonian of spin $S = 1/2$ frustrated XXZ Heisenberg chain in the presence of a staggered magnetic field. The amplitude of this field is proportional to the product of the parameter λ quantifying the broken translational symmetry of the underlying fermion model, and the spin-dependent hopping asymmetry parameter $t_{\uparrow} - t_{\downarrow}$. Thus, in contrast with the spin-isotropic case ($t_{\uparrow} = t_{\downarrow}$), the infrared properties of the spin-asymmetric model are described by a Hamiltonian with an explicitly broken translational symmetry.

It is instructive to check several limiting cases. In the case of spin-symmetric electron hopping ($t_{\uparrow} = t_{\downarrow} = t$), the effective Hamiltonian (56) reduces to the Hamiltonian of the isotropic ($SU(2)$ -invariant) Heisenberg chain

$$\mathcal{H}_{\text{eff}}^{(2)} = J \sum_n \mathbf{S}_n \cdot \mathbf{S}_{n+1}, \quad (58)$$

with a uniform exchange constant $J = 4t^2/U(1-\lambda^2)$. Thus, even if the translational symmetry of the underlying fermion model is broken ($\lambda \neq 0$), the second-order effective spin Hamiltonian remains translationally invariant.

In the complementary case of the Hubbard model with spin-dependent hopping ($\lambda = 0$, $t_{\uparrow} \neq t_{\downarrow}$), the second-order effective Hamiltonian properly reflects the broken spin symmetry and is given by the Hamiltonian of anisotropic ($U(1)$ -invariant) Heisenberg chain

$$\mathcal{H}_{\text{eff}}^{(2)} = J \sum_n (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y + \gamma S_n^z S_{n+1}^z), \quad (59)$$

with the anisotropy parameter $\gamma = (t_{\uparrow}^2 + t_{\downarrow}^2)/2t_{\uparrow}t_{\downarrow} > 1$.²²

Finally, in the limiting case of the Falicov–Kimball model ($t_{\downarrow} = 0$), the second-order effective spin Hamiltonian reduces to the Ising model in a staggered magnetic

field:

$$\mathcal{H}_{\text{eff}}^{(2)} = \sum_n (J_{\parallel} S_n^z S_{n+1}^z - (-1)^n h S_n^z), \quad (60)$$

where

$$J_{\parallel} = \frac{2t_{\uparrow}^2}{U(1-\lambda^2)}, \quad h = \frac{2\lambda t_{\uparrow}^2}{U(1-\lambda^2)}. \quad (61)$$

The physical mechanism responsible for the appearance of the staggered magnetic field in the effective spin Hamiltonian (56) can easily be traced in the ultimate limit of the Falicov–Kimball model, however, the argument remains valid also for arbitrary $t_{\uparrow} > t_{\downarrow} > 0$. Due to the doubling of the lattice unit cell, energetically it is preferable to locate all immobile fermions on odd sites, while the mobile up-spin fermions will dominantly occupy even sites. In this limit, the process of creation and annihilation of a doublon takes place only on odd sites and gives rise to the following Ising-type spin exchange parameter $J_{\parallel}^{(1)} = 2t_{\uparrow}^2/(U_o - \Delta)$, while in the opposite case, where all immobile spins are located on even sites, the same process yields the exchange constant $J_{\parallel}^{(2)} = 2t_{\uparrow}^2/(U_e + \Delta)$. The difference between the exchange energies for these two patterns equals

$$J_{\parallel}^{(1)} - J_{\parallel}^{(2)} = \frac{4\lambda t_{\uparrow}^2}{U(1-\lambda^2)} = 2h. \quad (62)$$

5.2. The fourth-order terms

The same technique as the one employed in the previous section can be used to rewrite the products of four T -terms in the effective Hamiltonian (47) via the spin $S = 1/2$ operators. There are 18 terms of this type. It is convenient to unite them in groups characterized by the similarity of the hopping processes and by the number of created doublons at the intermediate steps.

5.2.1. Group A: Four- T product terms of the form $T_{-1}T_0T_0T_1$

There are eight terms of this type in the effective Hamiltonian (47). In these processes the number of created doubly occupied sites is one. Four terms correspond to processes where the doublon is created and eventually annihilated on the same site, while the other four terms describe processes where the doublon is created on an odd (even) site and annihilated on the neighboring even (odd) site. The calculations are straightforward and one obtains the following expressions for the operators (the details can be found in the Appendix):

$$\begin{aligned} T_{-1}^o T_0^{\text{po}} T_0^{\text{pe}} T_1^o &= \sum_n \left\{ -t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2) (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) - 2t_{\uparrow}^2 t_{\downarrow}^2 \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) \right. \\ &\quad \left. + \frac{t_{\uparrow}^4 - t_{\downarrow}^4}{2} (-1)^n S_n^z - 2(t_{\uparrow}^4 - t_{\downarrow}^4) \left(S_{2n-1}^z S_{2n}^z - \frac{1}{4} \right) S_{2n+1}^z \right\} \end{aligned}$$

$$\begin{aligned}
& - (t_{\uparrow}^2 - t_{\downarrow}^2)^2 \left(S_{2n-1}^z S_{2n+1}^z - \frac{1}{4} \right) - 2t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 - t_{\downarrow}^2) [(S_{2n-1}^x S_{2n}^x \\
& + S_{2n-1}^y S_{2n}^y) S_{2n+1}^z + S_{2n-1}^z (S_{2n}^x S_{2n+1}^x + S_{2n}^y S_{2n+1}^y)] \Big\}, \quad (63)
\end{aligned}$$

$$\begin{aligned}
T_{-1}^o T_0^{\text{do}} T_0^{\text{de}} T_1^o &= \sum_n \left\{ -t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2) (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) - 2t_{\uparrow}^2 t_{\downarrow}^2 \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) \right. \\
& + \frac{t_{\uparrow}^4 - t_{\downarrow}^4}{2} (-1)^n S_n^z + 2(t_{\uparrow}^4 - t_{\downarrow}^4) \left(S_{2n}^z S_{2n+1}^z - \frac{1}{4} \right) S_{2n+2}^z \\
& - (t_{\uparrow}^2 - t_{\downarrow}^2)^2 \left(S_{2n}^z S_{2n+2}^z - \frac{1}{4} \right) + 2t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 - t_{\downarrow}^2) [(S_{2n}^x S_{2n+1}^x \\
& + S_{2n}^y S_{2n+1}^y) S_{2n+2}^z + S_{2n}^z (S_{2n+1}^x S_{2n+2}^x + S_{2n+1}^y S_{2n+2}^y)] \Big\}, \quad (64)
\end{aligned}$$

$$\begin{aligned}
T_{-1}^e T_0^{\text{pe}} T_0^{\text{po}} T_1^e &= \sum_n \left\{ -t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2) (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) - 2t_{\uparrow}^2 t_{\downarrow}^2 \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) \right. \\
& - \frac{t_{\uparrow}^4 - t_{\downarrow}^4}{2} (-1)^n S_n^z - 2(t_{\uparrow}^4 - t_{\downarrow}^4) \left(S_{2n}^z S_{2n+1}^z - \frac{1}{4} \right) S_{2n+2}^z \\
& - (t_{\uparrow}^2 - t_{\downarrow}^2)^2 \left(S_{2n}^z S_{2n+2}^z - \frac{1}{4} \right) - 2t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 - t_{\downarrow}^2) [(S_{2n}^x S_{2n+1}^x \\
& + S_{2n}^y S_{2n+1}^y) S_{2n+2}^z + S_{2n}^z (S_{2n+1}^x S_{2n+2}^x + S_{2n+1}^y S_{2n+2}^y)] \Big\}, \quad (65)
\end{aligned}$$

$$\begin{aligned}
T_{-1}^e T_0^{\text{de}} T_0^{\text{do}} T_1^e &= \sum_n \left\{ -t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2) (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) - 2t_{\uparrow}^2 t_{\downarrow}^2 \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) \right. \\
& - \frac{t_{\uparrow}^4 - t_{\downarrow}^4}{2} (-1)^n S_n^z + 2(t_{\uparrow}^4 - t_{\downarrow}^4) \left(S_{2n-1}^z S_{2n}^z - \frac{1}{4} \right) S_{2n+1}^z \\
& - (t_{\uparrow}^2 - t_{\downarrow}^2)^2 \left(S_{2n-1}^z S_{2n+1}^z - \frac{1}{4} \right) + 2t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 - t_{\downarrow}^2) [(S_{2n-1}^x S_{2n}^x \\
& + S_{2n-1}^y S_{2n}^y) S_{2n+1}^z + S_{2n-1}^z (S_{2n}^x S_{2n+1}^x + S_{2n}^y S_{2n+1}^y)] \Big\}, \quad (66)
\end{aligned}$$

$$\begin{aligned}
T_{-1}^e T_0^{\text{de}} T_0^{\text{pe}} T_1^o &= \sum_n \left\{ -t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2) (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) - 2t_{\uparrow}^2 t_{\downarrow}^2 \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) \right. \\
& + it_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 - t_{\downarrow}^2) [(S_{2n-1}^x S_{2n}^y - S_{2n-1}^y S_{2n}^x) \\
& - (S_{2n}^x S_{2n+1}^y - S_{2n}^y S_{2n+1}^x)] + 2t_{\uparrow}^2 t_{\downarrow}^2 \left(\mathbf{S}_{2n-1} \cdot \mathbf{S}_{2n+1} - \frac{1}{4} \right) \Big\}, \quad (67)
\end{aligned}$$

$$\begin{aligned}
T_{-1}^e T_0^{\text{pe}} T_0^{\text{de}} T_1^o &= \sum_n \left\{ -t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2) (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) - 2t_{\uparrow}^2 t_{\downarrow}^2 \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) \right. \\
&\quad - it_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 - t_{\downarrow}^2) [(S_{2n}^x S_{2n+1}^y - S_{2n}^y S_{2n+1}^x) \\
&\quad \left. - (S_{2n+1}^x S_{2n+2}^y - S_{2n+1}^y S_{2n+2}^x)] + 2t_{\uparrow}^2 t_{\downarrow}^2 \left(\mathbf{S}_{2n} \cdot \mathbf{S}_{2n+2} - \frac{1}{4} \right) \right\}, \quad (68)
\end{aligned}$$

$$\begin{aligned}
T_{-1}^o T_0^{\text{do}} T_0^{\text{po}} T_1^e &= \sum_n \left\{ -t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2) (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) - 2t_{\uparrow}^2 t_{\downarrow}^2 \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) \right. \\
&\quad + it_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 - t_{\downarrow}^2) [(S_{2n}^x S_{2n+1}^y - S_{2n}^y S_{2n+1}^x) \\
&\quad \left. - (S_{2n+1}^x S_{2n+2}^y - S_{2n+1}^y S_{2n+2}^x)] + 2t_{\uparrow}^2 t_{\downarrow}^2 \left(\mathbf{S}_{2n} \cdot \mathbf{S}_{2n+2} - \frac{1}{4} \right) \right\}, \quad (69)
\end{aligned}$$

$$\begin{aligned}
T_{-1}^o T_0^{\text{po}} T_0^{\text{do}} T_1^e &= \sum_n \left\{ -t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2) (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) - 2t_{\uparrow}^2 t_{\downarrow}^2 \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) \right. \\
&\quad - it_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 - t_{\downarrow}^2) [(S_{2n-1}^x S_{2n}^y - S_{2n-1}^y S_{2n}^x) \\
&\quad \left. - (S_{2n}^x S_{2n+1}^y - S_{2n}^y S_{2n+1}^x)] + 2t_{\uparrow}^2 t_{\downarrow}^2 \left(\mathbf{S}_{2n-1} \cdot \mathbf{S}_{2n+1} - \frac{1}{4} \right) \right\}. \quad (70)
\end{aligned}$$

5.2.2. *Group B1: Four-T product terms of the form $T_{-1}^a T_{-1}^b T_1^a T_1^b$ and $T_{-1}^b T_{-1}^a T_1^b T_1^a$ ($a \neq b$)*

There are four terms of this type in the effective Hamiltonian (47). These terms correspond to processes where two doublons are created on even and odd sites and then consecutively annihilated. If the first pair is created on site ℓ , the hopping processes leading to the creation of the second pair are restricted by the existence of an empty site adjacent to ℓ . Consequently, using the relations (52) and (53) one obtains a restricted double summation of the form

$$\begin{aligned}
T_{-1}^o T_{-1}^e T_1^o T_1^e &= \sum_{n, m \neq n, n \pm 1} \left[-2t_{\uparrow} t_{\downarrow} (S_m^x S_{m+1}^x + S_m^y S_{m+1}^y) - (t_{\uparrow}^2 + t_{\downarrow}^2) \left(S_m^z S_{m+1}^z - \frac{1}{4} \right) \right. \\
&\quad \left. + \frac{t_{\uparrow}^2 - t_{\downarrow}^2}{2} (-1)^m (S_m^z - S_{m+1}^z) \right] \cdot \left[-2t_{\uparrow} t_{\downarrow} (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) \right. \\
&\quad \left. - (t_{\uparrow}^2 + t_{\downarrow}^2) \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) - \frac{t_{\uparrow}^2 - t_{\downarrow}^2}{2} (-1)^n (S_n^z - S_{n+1}^z) \right] \\
&\equiv (T_{-1}^o T_1^o) * (T_{-1}^e T_1^e). \quad (71)
\end{aligned}$$

Here, we have introduced the notation $*$ to denote multiplication of infinite sums over the indices n and m with the restrictive condition $m \neq n, n \pm 1$.

For the other terms of the same group, we analogously obtain

$$T_{-1}^e T_{-1}^o T_1^o T_1^e = (T_{-1}^o T_1^o) * (T_{-1}^e T_1^e), \quad (72)$$

$$T_{-1}^e T_{-1}^o T_1^e T_1^o = T_{-1}^o T_{-1}^e T_1^e T_1^o = (T_{-1}^e T_1^e) * (T_{-1}^o T_1^o). \quad (73)$$

However, since all the hopping processes in these products take place on disjoint pairs of sites $(n, n+1)$ and $(m, m+1)$, one can freely commute the S -operators past each other, so that the order of the multiplicands becomes irrelevant:

$$\begin{aligned} (T_{-1}^o T_1^o) * (T_{-1}^e T_1^e) &= (T_{-1}^e T_1^e) * (T_{-1}^o T_1^o) \\ &= \frac{1}{2} [(T_{-1}^o T_1^o) * (T_{-1}^e T_1^e) + (T_{-1}^e T_1^e) * (T_{-1}^o T_1^o)]. \end{aligned} \quad (74)$$

5.2.3. Group B2: Four- T product terms of the form $T_{-1}^a T_{-1}^a T_1^a T_1^a$

There are two terms of this type in the effective Hamiltonian (47). These terms correspond to processes where two doubly occupied sites are created either on even or on odd sites and then consecutively annihilated. As before, the creation of the first pair puts limitations on the processes responsible for the creation of the second pair. In addition, since there are two different ways how one can get the same configuration corresponding to the pair of doublons located on two odd or two even sites, an extra factor of 2 appears in the expressions for these terms:

$$\begin{aligned} T_{-1}^o T_{-1}^o T_1^o T_1^o &= 2 \sum_{n, m \neq n, n \pm 1} \left[-2t_{\uparrow} t_{\downarrow} (S_m^x S_{m+1}^x + S_m^y S_{m+1}^y) - (t_{\uparrow}^2 + t_{\downarrow}^2) \left(S_m^z S_{m+1}^z - \frac{1}{4} \right) \right. \\ &\quad + \frac{t_{\uparrow}^2 - t_{\downarrow}^2}{2} (-1)^m (S_m^z - S_{m+1}^z) \cdot [-2t_{\uparrow} t_{\downarrow} (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) \\ &\quad \left. - (t_{\uparrow}^2 + t_{\downarrow}^2) \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) + \frac{t_{\uparrow}^2 - t_{\downarrow}^2}{2} (-1)^n (S_n^z - S_{n+1}^z) \right] \\ &\equiv 2(T_{-1}^o T_1^o) * (T_{-1}^o T_1^o), \end{aligned} \quad (75)$$

$$\begin{aligned} T_{-1}^e T_{-1}^e T_1^e T_1^e &= 2 \sum_{n, m \neq n, n \pm 1} \left[-2t_{\uparrow} t_{\downarrow} (S_m^x S_{m+1}^x + S_m^y S_{m+1}^y) - (t_{\uparrow}^2 + t_{\downarrow}^2) \left(S_m^z S_{m+1}^z - \frac{1}{4} \right) \right. \\ &\quad - \frac{t_{\uparrow}^2 - t_{\downarrow}^2}{2} (-1)^m (S_m^z - S_{m+1}^z) \cdot [-2t_{\uparrow} t_{\downarrow} (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) \\ &\quad \left. - (t_{\uparrow}^2 + t_{\downarrow}^2) \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) - \frac{t_{\uparrow}^2 - t_{\downarrow}^2}{2} (-1)^n (S_n^z - S_{n+1}^z) \right] \\ &\equiv 2(T_{-1}^e T_1^e) * (T_{-1}^e T_1^e). \end{aligned} \quad (76)$$

5.2.4. Group C: Four- T product terms of the form $T_{-1}T_1T_{-1}T_1$

There are four terms of this type in the effective Hamiltonian (47). These terms correspond to processes where a doublon is created and immediately annihilated on a site ℓ and then another doublon is created and annihilated on an arbitrary site ℓ' . Using (52) and (53), we obtain

$$\begin{aligned} T_{-1}^o T_1^o T_{-1}^o T_1^o &= \sum_{n,m} \left[-2t_{\uparrow}t_{\downarrow}(S_m^x S_{m+1}^x + S_m^y S_{m+1}^y) - (t_{\uparrow}^2 + t_{\downarrow}^2) \left(S_m^z S_{m+1}^z - \frac{1}{4} \right) \right. \\ &\quad \left. + \frac{t_{\uparrow}^2 - t_{\downarrow}^2}{2} (-1)^m (S_m^z - S_{m+1}^z) \right] \cdot [-2t_{\uparrow}t_{\downarrow}(S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) \\ &\quad - (t_{\uparrow}^2 + t_{\downarrow}^2) \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) + \frac{t_{\uparrow}^2 - t_{\downarrow}^2}{2} (-1)^n (S_n^z - S_{n+1}^z)] \\ &\equiv (T_{-1}^o T_1^o) \cdot (T_{-1}^o T_1^o), \end{aligned} \quad (77)$$

$$\begin{aligned} T_{-1}^e T_1^e T_{-1}^e T_1^e &= \sum_{n,m} \left[-2t_{\uparrow}t_{\downarrow}(S_m^x S_{m+1}^x + S_m^y S_{m+1}^y) - (t_{\uparrow}^2 + t_{\downarrow}^2) \left(S_m^z S_{m+1}^z - \frac{1}{4} \right) \right. \\ &\quad \left. - \frac{t_{\uparrow}^2 - t_{\downarrow}^2}{2} (-1)^m (S_m^z - S_{m+1}^z) \right] \cdot \left[-2t_{\uparrow}t_{\downarrow}(S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) \right. \\ &\quad \left. - (t_{\uparrow}^2 + t_{\downarrow}^2) \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) - \frac{t_{\uparrow}^2 - t_{\downarrow}^2}{2} (-1)^n (S_n^z - S_{n+1}^z) \right] \\ &\equiv (T_{-1}^e T_1^e) \cdot (T_{-1}^e T_1^e) \end{aligned} \quad (78)$$

and

$$T_{-1}^o T_1^o T_{-1}^e T_1^e = (T_{-1}^o T_1^o) \cdot (T_{-1}^e T_1^e), \quad T_{-1}^e T_1^e T_{-1}^o T_1^o = (T_{-1}^e T_1^e) \cdot (T_{-1}^o T_1^o). \quad (79)$$

As we observe, in marked contrast with the first eight terms (63)–(70) which only couple spins located on neighboring sites, the remaining 10 terms given by (71)–(79) contain a countless number of all possible two-spin $S_n^z S_m^z$, three-spin $S_n^p S_{n+1}^p S_m^z$ and four-spin $S_n^p S_{n+1}^p S_m^q S_{m+1}^q$ combinations, where $p, q = x, y, z$. The situation is rescued by the fact that after combining identical terms in the Hamiltonian (47), each term of the type $(T_{-1}^a T_1^a) * (T_{-1}^b T_1^b)$ will have its counterpart of the type $(T_{-1}^a T_1^a) \cdot (T_{-1}^b T_1^b)$ with just the opposite coefficient. As a result, all terms corresponding to distant spin–spin interaction are canceled:

$$\begin{aligned} &(T_{-1}^o T_1^o) \cdot (T_{-1}^o T_1^o) - (T_{-1}^o T_1^o) * (T_{-1}^o T_1^o) \\ &= \sum_{n,m=n,n\pm 1} \left[-2t_{\uparrow}t_{\downarrow}(S_m^x S_{m+1}^x + S_m^y S_{m+1}^y) - (t_{\uparrow}^2 + t_{\downarrow}^2) \left(S_m^z S_{m+1}^z - \frac{1}{4} \right) \right. \\ &\quad \left. + \frac{t_{\uparrow}^2 - t_{\downarrow}^2}{2} (-1)^m (S_m^z - S_{m+1}^z) \right] \cdot \left[-2t_{\uparrow}t_{\downarrow}(S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) \right. \\ &\quad \left. - (t_{\uparrow}^2 + t_{\downarrow}^2) \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) + \frac{t_{\uparrow}^2 - t_{\downarrow}^2}{2} (-1)^n (S_n^z - S_{n+1}^z) \right] \end{aligned}$$

$$\begin{aligned}
&= \sum_n \left\{ \left[-4t_{\uparrow}t_{\downarrow}(t_{\uparrow}^2+t_{\downarrow}^2)(S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) - (3t_{\uparrow}^4 + 2t_{\uparrow}^2 t_{\downarrow}^2 + 3t_{\downarrow}^4) \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) \right. \right. \\
&\quad + 2t_{\uparrow}^2 t_{\downarrow}^2 (S_n^x S_{n+2}^x + S_n^y S_{n+2}^y) + (t_{\uparrow}^4 + t_{\downarrow}^4) \left(S_n^z S_{n+2}^z - \frac{1}{4} \right) \Big] \\
&\quad + (-1)^n \left[\frac{t_{\uparrow}^4 - t_{\downarrow}^4}{2} (4S_{n-1}^z S_n^z S_{n+1}^z + 5S_n^z) \right. \\
&\quad \left. \left. + 2t_{\uparrow}t_{\downarrow}(t_{\uparrow}^2 - t_{\downarrow}^2)[S_{n-1}^z (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) + (S_{n-1}^x S_n^x + S_{n-1}^y S_n^y) S_{n+1}^z] \right] \right\}, \tag{80}
\end{aligned}$$

$$\begin{aligned}
&(T_{-1}^e T_1^e) \cdot (T_{-1}^e T_1^e) - (T_{-1}^e T_1^e) * (T_{-1}^e T_1^e) \\
&= \sum_{n,m=n,n\pm 1} \left[-2t_{\uparrow}t_{\downarrow}(S_m^x S_{m+1}^x + S_m^y S_{m+1}^y) - (t_{\uparrow}^2 + t_{\downarrow}^2) \left(S_m^z S_{m+1}^z - \frac{1}{4} \right) \right. \\
&\quad \left. - \frac{t_{\uparrow}^2 - t_{\downarrow}^2}{2} (-1)^m (S_m^z - S_{m+1}^z) \right] \cdot \left[-2t_{\uparrow}t_{\downarrow}(S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) \right. \\
&\quad \left. - (t_{\uparrow}^2 + t_{\downarrow}^2) \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) - \frac{t_{\uparrow}^2 - t_{\downarrow}^2}{2} (-1)^n (S_n^z - S_{n+1}^z) \right] \\
&= \sum_n \left\{ \left[-4t_{\uparrow}t_{\downarrow}(t_{\uparrow}^2 + t_{\downarrow}^2)(S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) - (3t_{\uparrow}^4 + 2t_{\uparrow}^2 t_{\downarrow}^2 + 3t_{\downarrow}^4) \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) \right. \right. \\
&\quad + 2t_{\uparrow}^2 t_{\downarrow}^2 (S_n^x S_{n+2}^x + S_n^y S_{n+2}^y) + (t_{\uparrow}^4 + t_{\downarrow}^4) \left(S_n^z S_{n+2}^z - \frac{1}{4} \right) \Big] \\
&\quad - (-1)^n \left[\frac{t_{\uparrow}^4 - t_{\downarrow}^4}{2} (4S_{n-1}^z S_n^z S_{n+1}^z + 5S_n^z) \right. \\
&\quad \left. \left. + 2t_{\uparrow}t_{\downarrow}(t_{\uparrow}^2 - t_{\downarrow}^2)[S_{n-1}^z (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) + (S_{n-1}^x S_n^x + S_{n-1}^y S_n^y) S_{n+1}^z] \right] \right\} \tag{81}
\end{aligned}$$

and

$$\begin{aligned}
&\frac{1}{2}[(T_{-1}^o T_1^o) \cdot (T_{-1}^e T_1^e) - (T_{-1}^o T_1^o) * (T_{-1}^e T_1^e)] \\
&\quad + \frac{1}{2}[(T_{-1}^e T_1^e) \cdot (T_{-1}^o T_1^o) - (T_{-1}^e T_1^e) * (T_{-1}^o T_1^o)] \\
&= \sum_n \left[-4t_{\uparrow}t_{\downarrow}(t_{\uparrow}^2 + t_{\downarrow}^2)(S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) - 8t_{\uparrow}^2 t_{\downarrow}^2 \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) \right. \\
&\quad \left. + 2t_{\uparrow}^2 t_{\downarrow}^2 (S_n^x S_{n+2}^x + S_n^y S_{n+2}^y) + 2t_{\uparrow}^2 t_{\downarrow}^2 \left(S_n^z S_{n+2}^z - \frac{1}{4} \right) \right]. \tag{82}
\end{aligned}$$

For the last two terms we have taken their combination to avoid calculation of extra terms which cancel each other in the sum.

Inserting finally the relations (63)–(82) into (47), we obtain that up to the fourth order the strong-coupling effective spin Hamiltonian for the spin-asymmetric alternating- U IHM is given by

$$\begin{aligned}
\mathcal{H}_{\text{eff}}^{(4)} = & -\frac{T_{-1}^o T_1^o}{U_o - \Delta} - \frac{T_{-1}^e T_1^e}{U_e + \Delta} - \frac{1}{U_o} \left[\frac{T_{-1}^o T_0^{\text{po}} T_0^{\text{pe}} T_1^o}{(U_o - \Delta)^2} + \frac{T_{-1}^e T_0^{\text{de}} T_0^{\text{do}} T_1^e}{(U_e + \Delta)^2} \right] \\
& - \frac{1}{U_e} \left[\frac{T_{-1}^o T_0^{\text{do}} T_0^{\text{de}} T_1^o}{(U_o - \Delta)^2} + \frac{T_{-1}^e T_0^{\text{po}} T_0^{\text{pe}} T_1^e}{(U_e + \Delta)^2} \right] \\
& - \frac{1}{(U_o - \Delta)U_o(U_e + \Delta)} [T_{-1}^o T_0^{\text{po}} T_0^{\text{do}} T_1^e + T_{-1}^e T_0^{\text{de}} T_0^{\text{pe}} T_1^o] \\
& - \frac{1}{(U_o - \Delta)U_e(U_e + \Delta)} [T_{-1}^o T_0^{\text{do}} T_0^{\text{po}} T_1^e + T_{-1}^e T_0^{\text{pe}} T_0^{\text{de}} T_1^o] \\
& + \frac{1}{(U_o - \Delta)^3} [(T_{-1}^o T_1^o) \cdot (T_{-1}^o T_1^o) - (T_{-1}^o T_1^o) * (T_{-1}^o T_1^o)] \\
& + \frac{1}{(U_e + \Delta)^3} [(T_{-1}^e T_1^e) \cdot (T_{-1}^e T_1^e) - (T_{-1}^e T_1^e) * (T_{-1}^e T_1^e)] \\
& + \frac{U_o + U_e}{(U_o - \Delta)^2 (U_e + \Delta)^2} \left\{ \frac{1}{2} [(T_{-1}^o T_1^o) \cdot (T_{-1}^e T_1^e) - (T_{-1}^o T_1^o) * (T_{-1}^e T_1^e)] \right. \\
& \left. + \frac{1}{2} [(T_{-1}^e T_1^e) \cdot (T_{-1}^o T_1^o) - (T_{-1}^e T_1^e) * (T_{-1}^o T_1^o)] \right\} \\
= & \sum_n \left\{ J_{\perp} (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) + J_{\parallel} \left(S_n^z S_{n+1}^z - \frac{1}{4} \right) \right. \\
& + J'_{\perp}(n) (S_n^x S_{n+2}^x + S_n^y S_{n+2}^y) + J'_{\parallel}(n) \left(S_n^z S_{n+2}^z - \frac{1}{4} \right) \\
& + W_{\perp}(n) [(S_{n-1}^x S_n^x + S_{n-1}^y S_n^y) S_{n+1}^z + S_{n-1}^z (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y)] \\
& \left. + W_{\parallel}(n) S_{n-1}^z S_n^z S_{n+1}^z - h(n) S_n^z \right\}, \tag{83}
\end{aligned}$$

where

$$\begin{aligned}
J_{\perp} = & 2t_{\uparrow} t_{\downarrow} \left[\frac{1}{U_o - \Delta} + \frac{1}{U_e + \Delta} \right] \\
& + t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2) \frac{U_o + U_e}{U_o U_e} \left[\frac{1}{(U_o - \Delta)^2} + \frac{1}{(U_e + \Delta)^2} + \frac{2}{(U_o - \Delta)(U_e + \Delta)} \right] \\
& - 4t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2) \left[\frac{1}{(U_o - \Delta)^3} + \frac{1}{(U_e + \Delta)^3} + \frac{U_o + U_e}{(U_o - \Delta)^2 (U_e + \Delta)^2} \right] \\
= & \frac{4t_{\uparrow} t_{\downarrow}}{U} \frac{1}{1 - \lambda^2} + \frac{t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2)}{U^3} \frac{2}{(1 - \delta^2)} \frac{4}{(1 - \lambda^2)^2} - \frac{4t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2)}{U^3} \frac{4(1 + \lambda^2)}{(1 - \lambda^2)^3}
\end{aligned}$$

$$= \frac{4t_{\uparrow}t_{\downarrow}}{U(1-\lambda^2)} \left[1 - \frac{2(t_{\uparrow}^2 + t_{\downarrow}^2)}{U^2(1-\lambda^2)^2} \left(2 + 2\lambda^2 - \frac{1-\lambda^2}{1-\delta^2} \right) \right], \quad (84)$$

$$\begin{aligned} J_{\parallel} &= (t_{\uparrow}^2 + t_{\downarrow}^2) \left[\frac{1}{U_o - \Delta} + \frac{1}{U_e + \Delta} \right] - 3(t_{\uparrow}^4 + t_{\downarrow}^4) \left[\frac{1}{(U_o - \Delta)^3} + \frac{1}{(U_e + \Delta)^3} \right] \\ &\quad + 2t_{\uparrow}^2 t_{\downarrow}^2 \frac{U_o + U_e}{U_o U_e} \left[\frac{1}{(U_o - \Delta)^2} + \frac{1}{(U_e + \Delta)^2} + \frac{2}{(U_o - \Delta)(U_e + \Delta)} \right] \\ &\quad - 2t_{\uparrow}^2 t_{\downarrow}^2 \left[\frac{1}{(U_o - \Delta)^3} + \frac{1}{(U_e + \Delta)^3} + \frac{4(U_o + U_e)}{(U_o - \Delta)^2 (U_e + \Delta)^2} \right] \\ &= \frac{2(t_{\uparrow}^2 + t_{\downarrow}^2)}{U(1-\lambda^2)} - \frac{6(t_{\uparrow}^4 + t_{\downarrow}^4)}{U^3(1-\lambda^2)^3} (1 + 3\lambda^2) - \frac{4t_{\uparrow}^2 t_{\downarrow}^2}{U^3(1-\lambda^2)^3} \left(5 - \lambda^2 - \frac{4(1-\lambda^2)}{1-\delta^2} \right), \end{aligned} \quad (85)$$

$$\begin{aligned} J'_{\perp}(n) &= -2t_{\uparrow}^2 t_{\downarrow}^2 \frac{4}{[(U_e + U_o) + (-1)^n (U_e - U_o)](U_o - \Delta)(U_e + \Delta)} \\ &\quad + 2t_{\uparrow}^2 t_{\downarrow}^2 \left[\frac{1}{(U_o - \Delta)^3} + \frac{1}{(U_e + \Delta)^3} + \frac{U_e + U_o}{(U_o - \Delta)^2 (U_e + \Delta)^2} \right] \\ &= \frac{4t_{\uparrow}^2 t_{\downarrow}^2}{U^3(1-\lambda^2)^3} \left[2 + 2\lambda^2 - \frac{(1-\lambda^2)^2}{1-\delta^2} \right] + (-1)^n \frac{4\delta t_{\uparrow}^2 t_{\downarrow}^2}{U^3(1-\lambda^2)(1-\delta^2)}, \end{aligned} \quad (86)$$

$$\begin{aligned} J'_{\parallel}(n) &= (t_{\uparrow}^2 - t_{\downarrow}^2)^2 \frac{2}{(U_e + U_o) + (-1)^n (U_e - U_o)} \left[\frac{1}{(U_o - \Delta)^2} + \frac{1}{(U_e + \Delta)^2} \right] \\ &\quad - 2t_{\uparrow}^2 t_{\downarrow}^2 \frac{4}{[(U_e + U_o) + (-1)^n (U_e - U_o)](U_o - \Delta)(U_e + \Delta)} \\ &\quad + (t_{\uparrow}^4 + t_{\downarrow}^4) \left[\frac{1}{(U_o - \Delta)^3} + \frac{1}{(U_e + \Delta)^3} \right] + 2t_{\uparrow}^2 t_{\downarrow}^2 \frac{U_e + U_o}{(U_o - \Delta)^2 (U_e + \Delta)^2} \\ &= -\frac{4t_{\uparrow}^2 t_{\downarrow}^2 (1 + \delta^2)}{U^3(1-\lambda^2)^2 (1-\delta^2)} + \frac{2(t_{\uparrow}^4 + t_{\downarrow}^4)}{U^3(1-\lambda^2)^3} \left[1 + 3\lambda^2 + \frac{1-\lambda^4}{1-\delta^2} \right] \\ &\quad + (-1)^n \frac{2\delta}{U^3(1-\lambda^2)^2 (1-\delta^2)} [4t_{\uparrow}^2 t_{\downarrow}^2 - (t_{\uparrow}^4 + t_{\downarrow}^4)(1 + \lambda^2)], \end{aligned} \quad (87)$$

$$\begin{aligned} W_{\perp}(n) &= (-1)^n \left\{ 2t_{\uparrow}t_{\downarrow}(t_{\uparrow}^2 - t_{\downarrow}^2) \frac{2}{(U_e + U_o) - (-1)^n (U_e - U_o)} \left[\frac{1}{(U_o - \Delta)^2} \right. \right. \\ &\quad \left. \left. - \frac{1}{(U_e + \Delta)^2} \right] + 2t_{\uparrow}t_{\downarrow}(t_{\uparrow}^2 - t_{\downarrow}^2) \left[\frac{1}{(U_o - \Delta)^3} - \frac{1}{(U_e + \Delta)^3} \right] \right\} \\ &= \frac{4\lambda t_{\uparrow}t_{\downarrow}(t_{\uparrow}^2 - t_{\downarrow}^2)}{U^3(1-\lambda^2)^2} \left\{ \frac{2\delta}{1-\delta^2} + (-1)^n \left[\frac{3+\lambda^2}{1-\lambda^2} + \frac{2}{1-\delta^2} \right] \right\}, \end{aligned} \quad (88)$$

$$\begin{aligned}
W_{\parallel}(n) &= (-1)^n \left\{ 2(t_{\uparrow}^4 - t_{\downarrow}^4) \frac{2}{(U_e + U_o) - (-1)^n(U_e - U_o)} \left[\frac{1}{(U_o - \Delta)^2} \right. \right. \\
&\quad \left. \left. - \frac{1}{(U_e + \Delta)^2} \right] + 2(t_{\uparrow}^4 - t_{\downarrow}^4) \left[\frac{1}{(U_o - \Delta)^3} - \frac{1}{(U_e + \Delta)^3} \right] \right\} \\
&= \frac{4\lambda(t_{\uparrow}^4 - t_{\downarrow}^4)}{U^3(1 - \lambda^2)^2} \left\{ \frac{2\delta}{1 - \delta^2} + (-1)^n \left[\frac{3 + \lambda^2}{1 - \lambda^2} + \frac{2}{1 - \delta^2} \right] \right\} \quad (89)
\end{aligned}$$

$$\begin{aligned}
h(n) &= (-1)^n \left\{ (t_{\uparrow}^2 - t_{\downarrow}^2) \left[\frac{1}{U_o - \Delta} - \frac{1}{U_e + \Delta} \right] \right. \\
&\quad + \frac{(t_{\uparrow}^4 - t_{\downarrow}^4)}{2} \frac{2}{(U_e + U_o) - (-1)^n(U_e - U_o)} \left[\frac{1}{(U_o - \Delta)^2} - \frac{1}{(U_e + \Delta)^2} \right] \\
&\quad \left. - \frac{5(t_{\uparrow}^4 - t_{\downarrow}^4)}{2} \left[\frac{1}{(U_o - \Delta)^3} - \frac{1}{(U_e + \Delta)^3} \right] \right\} \\
&= (-1)^n \left\{ \frac{2\lambda(t_{\uparrow}^2 - t_{\downarrow}^2)}{U(1 - \lambda^2)} - \frac{\lambda(t_{\uparrow}^4 - t_{\downarrow}^4)}{U^3(1 - \lambda^2)^3} \left[5(3 + \lambda^2) - \frac{2(1 - \lambda^2)}{1 - \delta^2} \right] \right\} \\
&\quad + \frac{2\lambda\delta(t_{\uparrow}^4 - t_{\downarrow}^4)}{U^3(1 - \lambda^2)^2(1 - \delta^2)}. \quad (90)
\end{aligned}$$

It should be pointed out that the presented expressions (83)–(90) fully agree with the known results in the limiting cases of the standard⁹ ($t_{\uparrow} = t_{\downarrow} = t$, $\Delta = \delta = \lambda = 0$) and the alternating- U ¹⁶ ($t_{\uparrow} = t_{\downarrow} = t$, $\Delta = 0$, $\lambda = \delta \neq 0$) Hubbard models, but some of the fourth-order coefficients do not coincide with the results obtained previously for the ionic¹² ($t_{\uparrow} = t_{\downarrow} = t$, $\delta = 0$, $\lambda = \Delta/U = x$) and the spin-asymmetric²² ($t_{\uparrow} \neq t_{\downarrow}$, $\Delta = \delta = \lambda = 0$) Hubbard models. More specifically, for the ionic Hubbard chain we arrive at a different expression in the numerator of the nearest-neighbor coupling J , whereas for the spin-asymmetric Hubbard model the disparities concern the numerators of the coefficients J_{\parallel} and J'_{\parallel} . We presume that these discrepancies are due to the perturbative schemes adopted by the authors of Refs. 12 and 22, as it is known that some of these procedures are not sufficiently well-controlled at higher orders.^{66,67}

6. Conclusion

We have derived the effective spin Hamiltonian for the low-energy sector of the one-dimensional half-filled spin-asymmetric alternating- U IHM in the limit of strong on-site repulsion. The obtained Hamiltonian is that of a frustrated Heisenberg chain with alternating NNN exchange and three-spin coupling in the presence of a uniform and a staggered magnetic field. As expected, the NNN exchange is larger for two spins separated by a site with low on-site repulsion than for spins separated by a site with high on-site repulsion. The intensity of the three-spin coupling and the

amplitudes of the magnetic fields are proportional to the product of the parameter λ , which reflects the broken translational symmetry of the lattice, and the difference between up- and down-spin electron hopping amplitudes $t_\uparrow - t_\downarrow$. The most dominant effect, however, comes from the staggered magnetic field, and therefore, in marked contrast with the spin-isotropic case $t_\uparrow = t_\downarrow$, the ground state properties of the considered electron system are described by a spin-chain model with explicitly broken translational symmetry.

We also remark that the general picture outlined above remains valid in the case of a half-filled bipartite lattice of a higher dimension — to the lowest order one again obtains the anisotropic nearest-neighbor spin exchange and the staggered magnetic field which, as before, dominates any higher order terms arising from the more complex lattice geometry.

Acknowledgments

The authors would like to thank D Baeriswyl for careful reading of the manuscript and many useful comments and suggestions. We acknowledge support from the Swiss National Science Foundation through the SCOPES Grant No. IZ73Z0-128058. GIJ also acknowledges support from the Georgian National Science Foundation and Science and Technology Center in Ukraine through the joint Grant No. STCU-5906.

Appendix A. Derivation of the Effective Spin Exchange Expressions for the Four- T Product Terms of the Form $T_{-1}T_0T_0T_1$

In order to rewrite the products of four T -operators in the language of spin $S = 1/2$ operators, once again we make use of the Hubbard X -operators defined in Sec. 4; the procedure is in essence the same as the one employed in Sec. 5.1 for the terms consisting of two T -operators.

Let us consider, for example, the term $T_{-1}^o T_0^{po} T_0^{pe} T_1^o$, which belongs to the group of the processes where the electron pair is created and annihilated on the same site. In the summations below, the square brackets around the lattice indices $[n, m, k]$ indicate that for an even site $2m$ its odd partners $2k + 1$ and $2n + 1$ represent neighboring sites. Thus, for a fixed m we have two possible sets: $k = m - 1$ and $n = m$, or $k = m$ and $n = m - 1$.

$$\begin{aligned}
& T_{-1}^o T_0^{po} T_0^{pe} T_1^o \\
&= \sum_{[n,m,k]} \sum_{\sigma} \sigma t_{\sigma} X_{2m}^{\sigma 0} X_{2k+1}^{\bar{\sigma} 2} \cdot \sum_{\beta} t_{\beta}^2 X_{2n+1}^{\beta 0} X_{2m}^{0\beta} X_{2m}^{\beta 0} X_{2n+1}^{0\beta} \cdot \sum_{\alpha} \alpha t_{\alpha} X_{2k+1}^{2\bar{\alpha}} X_{2m}^{0\alpha} \\
&= \sum_{[n,m,k]} \sum_{\alpha, \beta, \sigma} \alpha \sigma t_{\alpha} t_{\sigma} t_{\beta}^2 X_{2k+1}^{\bar{\sigma} \bar{\alpha}} X_{2m}^{\sigma \alpha} X_{2n+1}^{\beta \beta} \\
&= \sum_{[n,m,k]} [t_{\uparrow}^2 X_{2k+1}^{\downarrow \downarrow} X_{2m}^{\uparrow \uparrow} + t_{\downarrow}^2 X_{2k+1}^{\uparrow \uparrow} X_{2m}^{\downarrow \downarrow} - t_{\uparrow} t_{\downarrow} (X_{2k+1}^{\uparrow \downarrow} X_{2m}^{\downarrow \uparrow} + X_{2k+1}^{\downarrow \uparrow} X_{2m}^{\uparrow \downarrow})]
\end{aligned}$$

$$\begin{aligned}
& \cdot [t_{\uparrow}^2 X_{2n+1}^{\uparrow\uparrow} + t_{\downarrow}^2 X_{2n+1}^{\downarrow\downarrow}] \\
&= \sum_{[n,m,k]} \left[(t_{\uparrow}^2 + t_{\downarrow}^2) \left(\frac{1}{4} - S_{2k+1}^z S_{2m}^z \right) + \frac{t_{\uparrow}^2 - t_{\downarrow}^2}{2} (S_{2m}^z - S_{2k+1}^z) \right. \\
&\quad \left. - t_{\uparrow} t_{\downarrow} (S_{2k+1}^+ S_{2m}^- + S_{2k+1}^- S_{2m}^+) \right] \cdot \left[\frac{t_{\uparrow}^2 + t_{\downarrow}^2}{2} + (t_{\uparrow}^2 - t_{\downarrow}^2) S_{2n+1}^z \right] \\
&= \sum_{[n,m,k]} \left[\frac{(t_{\uparrow}^2 + t_{\downarrow}^2)^2}{2} \left(\frac{1}{4} - S_{2k+1}^z S_{2m}^z \right) + \frac{(t_{\uparrow}^2 - t_{\downarrow}^2)^2}{2} \left(S_{2m}^z S_{2n+1}^z - \frac{1}{4} \right. \right. \\
&\quad \left. \left. + \frac{1}{4} - S_{2k+1}^z S_{2n+1}^z \right) + (t_{\uparrow}^4 - t_{\downarrow}^4) \left(\frac{1}{4} - S_{2k+1}^z S_{2m}^z \right) S_{2n+1}^z \right. \\
&\quad \left. + \frac{t_{\uparrow}^4 - t_{\downarrow}^4}{4} (S_{2m}^z - S_{2k+1}^z) - t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2) (S_{2k+1}^x S_{2m}^x + S_{2k+1}^y S_{2m}^y) \right. \\
&\quad \left. - 2t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 - t_{\downarrow}^2) (S_{2k+1}^x S_{2m}^x + S_{2k+1}^y S_{2m}^y) S_{2n+1}^z \right] \\
&= \sum_{\ell} 2t_{\uparrow}^2 t_{\downarrow}^2 \left(\frac{1}{4} - S_{\ell}^z S_{\ell+1}^z \right) + \sum_m (t_{\uparrow}^2 - t_{\downarrow}^2)^2 \left(\frac{1}{4} - S_{2m-1}^z S_{2m+1}^z \right) \\
&\quad + \sum_{\ell} \frac{t_{\uparrow}^4 - t_{\downarrow}^4}{2} (-1)^{\ell} S_{\ell}^z + \sum_m 2(t_{\uparrow}^4 - t_{\downarrow}^4) \left(\frac{1}{4} - S_{2m-1}^z S_{2m}^z \right) S_{2m+1}^z \\
&\quad - \sum_{\ell} t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2) (S_{\ell}^x S_{\ell+1}^x + S_{\ell}^y S_{\ell+1}^y) - \sum_m 2t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 - t_{\downarrow}^2) [(S_{2m-1}^x S_{2m}^x \\
&\quad + S_{2m-1}^y S_{2m}^y) S_{2m+1}^z + S_{2m-1}^z (S_{2m}^x S_{2m+1}^x + S_{2m}^y S_{2m+1}^y)]. \tag{A.1}
\end{aligned}$$

In a similar manner one can derive the expressions of the operators $T_{-1}^e T_0^{\text{pe}} T_0^{\text{po}} T_1^e$, $T_{-1}^o T_0^{\text{do}} T_0^{\text{de}} T_1^o$ and $T_{-1}^e T_0^{\text{de}} T_0^{\text{do}} T_1^e$, belonging to the same group as the operator considered above. However, it should be noted that one can obtain all of these expressions directly from (A.1) by switching the roles of the odd and the even sites and/or interchanging the hopping amplitudes $t_{\uparrow} \leftrightarrow t_{\downarrow}$:

$$\begin{aligned}
& T_{-1}^e T_0^{\text{pe}} T_0^{\text{po}} T_1^e \\
&= T_{-1}^o T_0^{\text{po}} T_0^{\text{pe}} T_1^o (2k+1, 2m, 2n+1 \rightarrow 2k, 2m+1, 2n) \\
&= \sum_{\ell} 2t_{\uparrow}^2 t_{\downarrow}^2 \left(\frac{1}{4} - S_{\ell}^z S_{\ell+1}^z \right) + \sum_m (t_{\uparrow}^2 - t_{\downarrow}^2)^2 \left(\frac{1}{4} - S_{2m}^z S_{2m+2}^z \right) \\
&\quad - \sum_{\ell} \frac{t_{\uparrow}^4 - t_{\downarrow}^4}{2} (-1)^{\ell} S_{\ell}^z + \sum_m 2(t_{\uparrow}^4 - t_{\downarrow}^4) \left(\frac{1}{4} - S_{2m}^z S_{2m+1}^z \right) S_{2m+2}^z
\end{aligned}$$

$$\begin{aligned}
& - \sum_{\ell} t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2) (S_{\ell}^x S_{\ell+1}^x + S_{\ell}^y S_{\ell+1}^y) - \sum_m 2t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 - t_{\downarrow}^2) [(S_{2m}^x S_{2m+1}^x \\
& + S_{2m}^y S_{2m+1}^y) S_{2m+2}^z + S_{2m}^z (S_{2m+1}^x S_{2m+2}^x + S_{2m+1}^y S_{2m+2}^y)], \quad (A.2)
\end{aligned}$$

$$\begin{aligned}
& T_{-1}^o T_0^{\text{do}} T_0^{\text{de}} T_1^o \\
& = T_{-1}^o T_0^{\text{po}} T_0^{\text{pe}} T_1^o (2k+1, 2m, 2n+1 \rightarrow 2k, 2m+1, 2n \text{ and } t_{\uparrow} \leftrightarrow t_{\downarrow}) \\
& = \sum_{\ell} 2t_{\uparrow}^2 t_{\downarrow}^2 \left(\frac{1}{4} - S_{\ell}^z S_{\ell+1}^z \right) + \sum_m (t_{\uparrow}^2 - t_{\downarrow}^2)^2 \left(\frac{1}{4} - S_{2m}^z S_{2m+2}^z \right) \\
& + \sum_{\ell} \frac{t_{\uparrow}^4 - t_{\downarrow}^4}{2} (-1)^{\ell} S_{\ell}^z - \sum_m 2(t_{\uparrow}^4 - t_{\downarrow}^4) \left(\frac{1}{4} - S_{2m}^z S_{2m+1}^z \right) S_{2m+2}^z \\
& - \sum_{\ell} t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2) (S_{\ell}^x S_{\ell+1}^x + S_{\ell}^y S_{\ell+1}^y) + \sum_m 2t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 - t_{\downarrow}^2) [(S_{2m}^x S_{2m+1}^x \\
& + S_{2m}^y S_{2m+1}^y) S_{2m+2}^z + S_{2m}^z (S_{2m+1}^x S_{2m+2}^x + S_{2m+1}^y S_{2m+2}^y)], \quad (A.3)
\end{aligned}$$

$$\begin{aligned}
& T_{-1}^e T_0^{\text{de}} T_0^{\text{do}} T_1^e \\
& = T_{-1}^o T_0^{\text{po}} T_0^{\text{pe}} T_1^o (t_{\uparrow} \leftrightarrow t_{\downarrow}) \\
& = \sum_{\ell} 2t_{\uparrow}^2 t_{\downarrow}^2 \left(\frac{1}{4} - S_{\ell}^z S_{\ell+1}^z \right) + \sum_m (t_{\uparrow}^2 - t_{\downarrow}^2)^2 \left(\frac{1}{4} - S_{2m-1}^z S_{2m+1}^z \right) \\
& - \sum_{\ell} \frac{t_{\uparrow}^4 - t_{\downarrow}^4}{2} (-1)^{\ell} S_{\ell}^z - \sum_m 2(t_{\uparrow}^4 - t_{\downarrow}^4) \left(\frac{1}{4} - S_{2m-1}^z S_{2m}^z \right) S_{2m+1}^z \\
& - \sum_{\ell} t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2) (S_{\ell}^x S_{\ell+1}^x + S_{\ell}^y S_{\ell+1}^y) + \sum_m 2t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 - t_{\downarrow}^2) [(S_{2m-1}^x S_{2m}^x \\
& + S_{2m-1}^y S_{2m}^y) S_{2m+1}^z + S_{2m-1}^z (S_{2m}^x S_{2m+1}^x + S_{2m}^y S_{2m+1}^y)]. \quad (A.4)
\end{aligned}$$

The expressions for the remaining four operators, corresponding to the processes where the pair is created on one site and annihilated on a neighboring one, can be established in an analogous way (utilizing where necessary the freedom of renaming the lattice indices $k \leftrightarrow n$ to facilitate the calculation):

$$\begin{aligned}
& T_{-1}^e T_0^{\text{de}} T_0^{\text{pe}} T_1^o \\
& = \sum_{[n,m,k]} \sum_{\sigma} \sigma t_{\sigma} X_{2n+1}^{\sigma 0} X_{2m}^{\bar{\sigma} 2} \cdot \sum_{\beta} t_{\bar{\beta}} t_{\beta} X_{2m}^{2\beta} X_{2k+1}^{\beta 2} X_{2m}^{\beta 0} X_{2n+1}^{0\beta} \cdot \sum_{\alpha} \alpha t_{\alpha} X_{2k+1}^{2\bar{\alpha}} X_{2m}^{0\alpha} \\
& = - \sum_{[n,m,k]} \sum_{\alpha, \beta, \sigma} \alpha \sigma t_{\alpha} t_{\sigma} t_{\bar{\beta}} t_{\beta} X_{2k+1}^{\beta \bar{\alpha}} X_{2m}^{\bar{\sigma} \alpha} X_{2n+1}^{\sigma \beta}
\end{aligned}$$

$$\begin{aligned}
&= \sum_{[n,m,k]} [-t_{\uparrow}^3 t_{\downarrow} (X_{2k+1}^{\uparrow\downarrow} X_{2m}^{\downarrow\uparrow} X_{2n+1}^{\uparrow\uparrow} + X_{2k+1}^{\downarrow\downarrow} X_{2m}^{\downarrow\uparrow} X_{2n+1}^{\uparrow\downarrow}) \\
&\quad - t_{\uparrow} t_{\downarrow}^3 (X_{2k+1}^{\uparrow\uparrow} X_{2m}^{\uparrow\downarrow} X_{2n+1}^{\downarrow\uparrow} + X_{2k+1}^{\downarrow\uparrow} X_{2m}^{\uparrow\downarrow} X_{2n+1}^{\downarrow\downarrow}) \\
&\quad + t_{\uparrow}^2 t_{\downarrow}^2 (X_{2k+1}^{\downarrow\uparrow} X_{2m}^{\downarrow\downarrow} X_{2n+1}^{\uparrow\downarrow} + X_{2k+1}^{\uparrow\downarrow} X_{2m}^{\uparrow\uparrow} X_{2n+1}^{\downarrow\uparrow} \\
&\quad + X_{2k+1}^{\downarrow\downarrow} X_{2m}^{\uparrow\uparrow} X_{2n+1}^{\downarrow\downarrow} + X_{2k+1}^{\uparrow\uparrow} X_{2m}^{\downarrow\downarrow} X_{2n+1}^{\uparrow\uparrow})] \\
&= \sum_{[n,m,k]} [-t_{\uparrow}^3 t_{\downarrow} X_{2k+1}^{\uparrow\downarrow} X_{2m}^{\downarrow\uparrow} - t_{\uparrow} t_{\downarrow}^3 X_{2k+1}^{\downarrow\uparrow} X_{2m}^{\uparrow\downarrow} \\
&\quad + t_{\uparrow}^2 t_{\downarrow}^2 (X_{2k+1}^{\uparrow\downarrow} X_{2n+1}^{\downarrow\uparrow} + X_{2k+1}^{\downarrow\downarrow} X_{2m}^{\uparrow\uparrow} X_{2n+1}^{\downarrow\downarrow} + X_{2k+1}^{\uparrow\uparrow} X_{2m}^{\downarrow\downarrow} X_{2n+1}^{\uparrow\uparrow})] \\
&= \sum_{[n,m,k]} \left[-t_{\uparrow}^3 t_{\downarrow} S_{2k+1}^+ S_{2m}^- - t_{\uparrow} t_{\downarrow}^3 S_{2k+1}^- S_{2m}^+ + t_{\uparrow}^2 t_{\downarrow}^2 \left[S_{2k+1}^+ S_{2n+1}^- \right. \right. \\
&\quad + \left(\frac{1}{2} - S_{2k+1}^z \right) \left(\frac{1}{2} + S_{2m}^z \right) \left(\frac{1}{2} - S_{2n+1}^z \right) \\
&\quad + \left. \left(\frac{1}{2} + S_{2k+1}^z \right) \left(\frac{1}{2} - S_{2m}^z \right) \left(\frac{1}{2} + S_{2n+1}^z \right) \right] \\
&= \sum_{[n,m,k]} \left[-t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2) (S_{2k+1}^x S_{2m}^x + S_{2k+1}^y S_{2m}^y) \right. \\
&\quad + i t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 - t_{\downarrow}^2) (S_{2k+1}^x S_{2m}^y - S_{2k+1}^y S_{2m}^x) \\
&\quad + t_{\uparrow}^2 t_{\downarrow}^2 \left[S_{2k+1}^+ S_{2n+1}^- + \left(\frac{1}{4} - S_{2k+1}^z S_{2m}^z \right) \right. \\
&\quad + \left. \left(\frac{1}{4} - S_{2m}^z S_{2n+1}^z \right) + \left(S_{2k+1}^z S_{2n+1}^z - \frac{1}{4} \right) \right] \\
&= - \sum_{\ell} t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2) (S_{\ell}^x S_{\ell+1}^x + S_{\ell}^y S_{\ell+1}^y) \\
&\quad + \sum_m i t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 - t_{\downarrow}^2) [S_{2m-1}^x S_{2m}^y - S_{2m-1}^y S_{2m}^x - (S_{2m}^x S_{2m+1}^y - S_{2m}^y S_{2m+1}^x)] \\
&\quad + \sum_m 2 t_{\uparrow}^2 t_{\downarrow}^2 (S_{2m-1}^x S_{2m+1}^x + S_{2m-1}^y S_{2m+1}^y) + \sum_{\ell} 2 t_{\uparrow}^2 t_{\downarrow}^2 \left(\frac{1}{4} - S_{\ell}^z S_{\ell+1}^z \right) \\
&\quad + \sum_m 2 t_{\uparrow}^2 t_{\downarrow}^2 \left(S_{2m-1}^z S_{2m+1}^z - \frac{1}{4} \right), \tag{A.5}
\end{aligned}$$

$$\begin{aligned}
&T_{-1}^{\circ} T_0^{\text{do}} T_0^{\text{po}} T_1^{\text{e}} \\
&= T_{-1}^{\text{e}} T_0^{\text{de}} T_0^{\text{pe}} T_1^{\circ} (2k+1, 2m, 2m+1 \rightarrow 2k, 2m+1, 2n)
\end{aligned}$$

$$\begin{aligned}
&= - \sum_{\ell} t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2) (S_{\ell}^x S_{\ell+1}^x + S_{\ell}^y S_{\ell+1}^y) \\
&\quad + \sum_m i t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 - t_{\downarrow}^2) [S_{2m}^x S_{2m+1}^y - S_{2m}^y S_{2m+1}^x - (S_{2m+1}^x S_{2m+2}^y - S_{2m+1}^y S_{2m+2}^x)] \\
&\quad + \sum_{\ell} 2t_{\uparrow}^2 t_{\downarrow}^2 \left(\frac{1}{4} - S_{\ell}^z S_{\ell+1}^z \right) + \sum_m 2t_{\uparrow}^2 t_{\downarrow}^2 \left(\mathbf{S}_{2m} \cdot \mathbf{S}_{2m+2} - \frac{1}{4} \right), \tag{A.6}
\end{aligned}$$

$$\begin{aligned}
&T_{-1}^o T_0^{\text{po}} T_0^{\text{do}} T_1^e \\
&= T_{-1}^e T_0^{\text{de}} T_0^{\text{pe}} T_1^o (t_{\uparrow} \leftrightarrow t_{\downarrow}) \\
&= - \sum_{\ell} t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2) (S_{\ell}^x S_{\ell+1}^x + S_{\ell}^y S_{\ell+1}^y) \\
&\quad - \sum_m i t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 - t_{\downarrow}^2) [S_{2m-1}^x S_{2m}^y - S_{2m-1}^y S_{2m}^x - (S_{2m}^x S_{2m+1}^y - S_{2m}^y S_{2m+1}^x)] \\
&\quad + \sum_{\ell} 2t_{\uparrow}^2 t_{\downarrow}^2 \left(\frac{1}{4} - S_{\ell}^z S_{\ell+1}^z \right) + \sum_m 2t_{\uparrow}^2 t_{\downarrow}^2 \left(\mathbf{S}_{2m-1} \cdot \mathbf{S}_{2m+1} - \frac{1}{4} \right), \tag{A.7}
\end{aligned}$$

$$\begin{aligned}
&T_{-1}^e T_0^{\text{pe}} T_0^{\text{de}} T_1^o \\
&= T_{-1}^e T_0^{\text{de}} T_0^{\text{pe}} T_1^o (2k+1, 2m, 2n+1 \rightarrow 2k, 2m+1, 2n \text{ and } t_{\uparrow} \leftrightarrow t_{\downarrow}) \\
&= - \sum_{\ell} t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 + t_{\downarrow}^2) (S_{\ell}^x S_{\ell+1}^x + S_{\ell}^y S_{\ell+1}^y) \\
&\quad - \sum_m i t_{\uparrow} t_{\downarrow} (t_{\uparrow}^2 - t_{\downarrow}^2) [S_{2m}^x S_{2m+1}^y - S_{2m}^y S_{2m+1}^x - (S_{2m+1}^x S_{2m+2}^y - S_{2m+1}^y S_{2m+2}^x)] \\
&\quad + \sum_{\ell} 2t_{\uparrow}^2 t_{\downarrow}^2 \left(\frac{1}{4} - S_{\ell}^z S_{\ell+1}^z \right) + \sum_m 2t_{\uparrow}^2 t_{\downarrow}^2 \left(\mathbf{S}_{2m} \cdot \mathbf{S}_{2m+2} - \frac{1}{4} \right). \tag{A.8}
\end{aligned}$$

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